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SCATTERING OF WAVES BY VISOELASTIC COMPOSITES AND THE CHARACTERIZATION OF THEIR DYNAMIC PROPERTIES

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MULTIPLE SCATTERING AND WAVES IN RANDOM MEDIA
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BULK PROPAGATION CHARACTERISTICS OF DISCRETE RANDOM MEDIA

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The propagation of electromagnetic waves in an infinite medium composed of a random distribution of identical, finite scatterers is studied. The T-matrix of a single isolated scatterer, obtained by using the null field equation, is used to make the equations for the field incident on a particular scatterer and the field scattered by it, self consistent. The method we propose is well suited for computations at wavelengths comparable to obstacle size and for non-spherical obstacles. The attenuation associated with the coherent field as predicted by our computations is compared with the only two sets of experimental results that can be found in the literature. Agreements and discrepancies are examined and the range of validity of the assumed quasi-crystalline approximation (~~QCA~~) is discussed. Further improvements using the coherent potential approximation (~~CPA~~) and the 'self consistent approximation' (~~SCA~~) as well as improved models of the pair correlation function are suggested.

1. INTRODUCTION

A treatment of the propagation of electromagnetic waves in an infinite medium composed of a random distribution of identical finite scatterers is presented. The coherent or average wave in such a medium will be assumed to be a plane wave propagating in a homogeneous continuum characterized by a "bulk" complex wave number. This wave number will depend on both the frequency and the concentration of the discrete scatterers causing the effective medium to be dispersive. The aim of this work is to present a multiple scattering theory and a computational method of obtaining such dispersion relations for random media models including comparisons with some past laboratory experiments.

The effects of multiple scattering on the coherent wave are of great practical importance, in particular the dependence on concentration at wavelengths comparable to scatterer size. At very low concentrations (< 1% by volume) multiple scattering can be neglected and each scatterer can be treated as independent. However, in many practical situations the concentration can range between 1% to 20% where multiple scattering effects may be important. This is particularly reflected in the study of higher order statistics of the random medium including radiative transfer theory which assume that the

coherent wave propagates with a wave number that is only dependent on the forward scattering amplitude of a single scatterer. This result is obtained as a solution of the Foldy-Twersky integral equation for an infinite slab medium filled with large tenuous scatterers [1]. However, this well known analysis neglects correlation between the scatterers, and recent advances in multiple scattering theory by Twersky [2,3,4] have accounted for the effects of pair-correlation at higher concentrations.

The theoretical/computational method presented here is based on a self-consistent exciting field approach and relies on the T-matrix [5] which characterizes the response of an isolated single scatterer to an arbitrary exciting field. The random medium is described statistically with respect to the random positions that each scatterer can occupy through the first and second order probability distribution functions. Ensemble or configurational averaging together with Lax's quasi-crystalline approximation (QCA) yields a set of "hole" correction integrals. By assuming a plane wave behavior for the coherent wave and using the extinction theorem gives rise to a homogeneous system of equations whose singular solutions yield the complex wave number. The method is necessarily computational; however, analytical forms of the dispersion relations are obtained in the low frequency or Rayleigh limit for spherical and spheroidal scatterer geometries. This paper closely follows the developments given by the authors [6,7,8] for acoustic, electromagnetic and elastic waves. Previous work which forms the basis for the present analysis is given in Refs. [9-12]. The numerical results obtained for a random distribution of spheres are compared with the experimental results of Hawley, Beard and Twersky [13] and Olsen and Kharadly [14].

One of the aims of this paper is to suggest improvements to the QCA by incorporating the coherent potential approximation (CPA). Certain multiple scattering processes that are neglected by the QCA-CPA scheme can be restored by making the 'self-consistent approximation' (SCA). If these improvements are used with more realistic forms of the pair correlation function, then it may be possible to extend the present formalism to a wider range of frequencies and concentrations.

2. MULTIPLE SCATTERING FORMALISM

Consider N identical scatterers located in free space as shown

in Fig. 1 where O_i and O_j refer to the centers of the i-th and j-th scatterers. The scatterers are assumed to be bodies of revolution with parallel symmetry axes, and the coherent wave is assume to propagate along this direction so that the bulk medium is isotropic and does not cause any depolarization. Let ε_r be the relative dielectric constant of the homogeneous scatterers.

We represent an incident plane electromagnetic wave propagating along the positive z axis with wave number k and of unit amplitude with an $e^{-i\omega t}$ time dependence by

$$\vec{E}^{o}(\vec{r}) = \hat{e}\exp(ikz) \tag{1}$$

where $\hat{e}$ is the unit polarization vector. With no loss of generality we choose $\hat{e}$ to be either $\hat{x}$ or $\hat{y}$.

The total electric field at any point in free space outside the scatterers is the sum of the incident field and the fields scattered by all the scatterers. This is written as

$$\vec{E}(\vec{r}) = \vec{E}^{o}(\vec{r}) + \sum_{i=1}^{N} \vec{E}_i^{s}(\vec{r} - \vec{r}_i) \quad . \tag{2}$$

where $\vec{E}_i^{s}(\vec{r} - \vec{r}_i)$ is the field, scattered by the i-th scatterer, at the point of observation $\vec{r}$. The field that excites the i-th scatterer, however, is the incident field plus the fields scattered from all the other scatterers. The exciting field term $\vec{E}^{e}$ is used to distinguish between the field actually incident on a scatterer and the external incident field, $\vec{E}^{o}$, produced by a source at infinity. Thus, at a point $\vec{r}$ in the vicinity of the i-th scatterer, we write

$$\vec{E}_i^{e}(\vec{r}) = \vec{E}^{o}(\vec{r}) + \sum_{j\neq i}^{N} \vec{E}_j^{s}(\vec{r} - \vec{r}_j) \ , \qquad a < |\vec{r} - \vec{r}_i| < 2a \tag{3}$$

where 'a' is the radius of the imaginary sphere circumscribing a scatterer (see Fig. 1). In this analysis, we have assumed that there is no interpenetration of the imaginary spherical shells of radius 'a' which circumscribe each scatterer.

The T-matrix formulation of scattering we adopt here is based on the extended integral equation approach due to Waterman [15]. The scattered and exciting fields with respect to a particular scatterer are expanded in terms of a complete set of basis functions $(M_{\sigma mn}, N_{\sigma mn})$ which are the vector spherical harmonics. These form

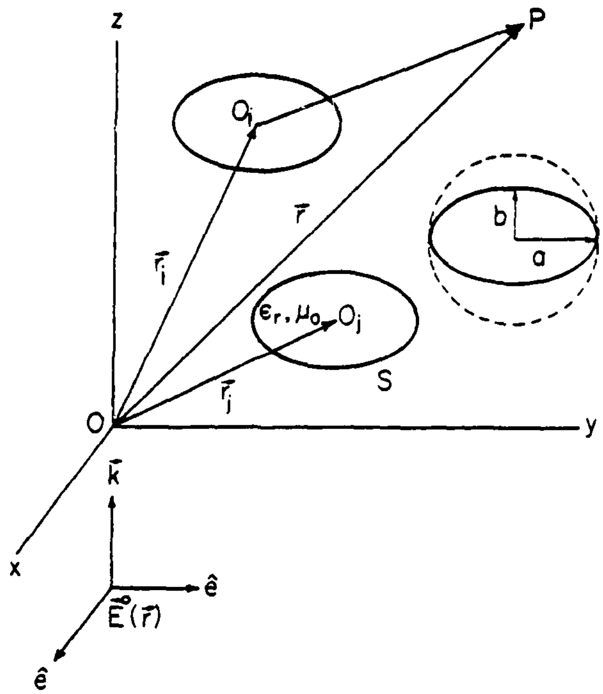

Figure 1. Random distribution of aligned scatterers and plane wave incidence in the z-direction

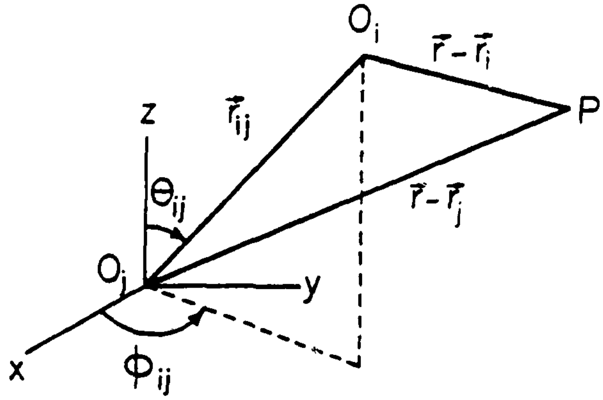

Figure 2. Geometry for the translation of the coordinate system from O_j to O_i

solutions to the vector Helmholtz equation and are given by

$$\vec{M}_{\sigma mn} = \nabla \times \left[\vec{r} h_n(kr)\ P_n^m(\cos\theta) \begin{matrix}\cos m\phi \\ \sin m\phi\end{matrix}\right] \tag{4a}$$

$$\vec{N}_{\sigma mn} = (1/k)\nabla \times \vec{M}_{\sigma mn} \tag{4b}$$

where $\sigma = \begin{matrix}e\\o\end{matrix}$ stands for even or odd and refers to the choice of the trigonometric functions in Eq. (4). Wave functions regular at the origin are obtained by replacing Hankel functions in Eq. (4) by the Bessel functions j_n and are denoted by $(Re\vec{M}_{\sigma mn}, Re\vec{N}_{\sigma mn})$.

The scattered field from, say, the j-th scatterer, $\vec{E}_j^s(\vec{r})$, can be expanded in terms of "outgoing" vector wave functions with unknown coefficients $(B^j_{\sigma\ell n}, C^j_{\sigma\ell n})$ as

$$\vec{E}_j^s(\vec{r}) = \sum_{n=0}^{\infty} \sum_{\ell=0}^{n} \sum_{\sigma=\begin{smallmatrix}e\\o\end{smallmatrix}} \left\{B^j_{\sigma\ell n}\ \vec{M}_{\sigma\ell n}(\vec{r}-\vec{r}_j) + C^j_{\sigma\ell n}\vec{N}_{\sigma\ell n}(\vec{r}-\vec{r}_j)\right\} ;$$

$$|\vec{r}-\vec{r}_j| > a\ . \tag{5}$$

The exciting field incident on, say, the i-th scatterer, $\vec{E}_i^e(\vec{r})$, can be expanded into regular wave functions with unknown coefficients $(b^i_{\sigma\ell n}, c^i_{\sigma\ell n})$ as

$$\vec{E}_i^e(\vec{r}) = \sum_{n=0}^{\infty} \sum_{\ell=0}^{n} \sum_{\sigma=\begin{smallmatrix}e\\o\end{smallmatrix}} \left\{b^i_{\sigma\ell n} Re\vec{M}_{\sigma\ell n}(\vec{r}-\vec{r}_i) + c^i_{\sigma\ell n} Re\vec{N}_{\sigma\ell n}(\vec{r}-\vec{r}_i)\right\};$$

$$a < |\vec{r}-\vec{r}_i| < 2a\ . \tag{6}$$

The choice of the basis set in Eq. (5) satisfies the radiation condition at infinity for the scattered field $\vec{E}_j^s$, while the choice in Eq. (6) is a result of the regular behavior of the exciting field $\vec{E}_i^e$ in the region $a < |\vec{r}-\vec{r}_i| < 2a$.

It has been shown that, if the total field outside a scatterer is the sum of incident and scattered fields, the unknown scattered field expansion coefficients can be related to the incident field expansion coefficients through the transition or T-matrix [5,15]. We extend this definition to the present case. Since $(\vec{E}_j^e + \vec{E}_j^s)$ is

the total field at any point in free space, the expansion coefficients of the field scattered by the j-th scatterer may be formally related to the coefficients of the field exciting the j-th scatterer through the T-matrix:

$$\begin{bmatrix} B^j_{\sigma\ell n} \\ C^j_{\sigma\ell n} \end{bmatrix} = \begin{bmatrix} \left(T^{\sigma\ell n}_{\tau mp}\right)^{11} & \left(T^{\sigma\ell n}_{\tau mp}\right)^{12} \\ \left(T^{\sigma\ell n}_{\tau mp}\right)^{21} & \left(T^{\sigma\ell n}_{\tau mp}\right)^{22} \end{bmatrix} \begin{bmatrix} b^j_{\tau mp} \\ c^j_{\tau mp} \end{bmatrix} \tag{7}$$

where summation is denoted by the repeated index convention. The elements of the T-matrix involve surface integrals, which can be evaluated in closed form for spherical geometry, while for scatterers of arbitrary shape they can only be evaluated numerically. The T-matrix for a single scatterer is of the form

$$T = (Q^{-1})\,\mathrm{Re}Q \tag{8}$$

where ReQ and Q are matrices which are functions of the surface S of the scatterer and of the nature of the boundary conditions.

Substitution of Eqs. (5) and (6) in Eq. (3) yields

$$\begin{aligned} \sum_{p=0}^{\infty} \sum_{m=0}^{p} \sum_{\tau=e,o} \left\{ b^i_{\tau mp} \mathrm{Re}\vec{M}_{\tau mp}(\vec{r}-\vec{r}_i) + c^i_{\tau mp} \mathrm{Re}\vec{N}_{\tau mp}(\vec{r}-\vec{r}_i) \right\} &= \vec{E}^o(\vec{r}) \\ + \sum_{j\neq i}^{N} \sum_{n=0}^{\infty} \sum_{\ell=0}^{n} \sum_{\sigma=e,o} \left\{ B^j_{\sigma\ell n} \vec{M}_{\sigma\ell n}(\vec{r}-\vec{r}_j) + C^j_{\sigma\ell n} \vec{N}_{\sigma\ell n}(\vec{r}-\vec{r}_j) \right\} . \end{aligned} \tag{9}$$

Note that the series on the right-hand side of Eq. (9) is expressed with respect to the center of the j-th scatterer while the series to the left is expressed with respect to the center of the i-th scatterer. The addition theorem for the vector basis functions will be used to express the right hand side of Eq. (9) in terms of the center of the i-th scatterer. Formulas for the translation of $\{\vec{M}(\vec{r}-\vec{r}_j), \vec{N}(\vec{r}-\vec{r}_j)\}$ to an origin centered around the i-th scatterer can be found in [16,17]:

$$\vec{M}_{\sigma\ell n}(\vec{r}-\vec{r}_j) = \sum_{n'=0}^{\infty}\sum_{m'=0}^{n}\sum_{\tau=e}^{o}\left[A^{\sigma\ell n}_{\tau m'n'}\ \mathrm{Re}\vec{M}_{\tau m'n'}(\vec{r}-\vec{r}_i)\right. \tag{10a}$$

$$\left. + B^{\sigma\ell n}_{\tau m'n'}\ \mathrm{Re}\vec{N}_{\tau m'n'}(\vec{r}-\vec{r}_i)\right] ,$$

$$\vec{N}_{\sigma\ell n}(\vec{r}-\vec{r}_j) = \sum_{n'=0}^{\infty}\sum_{m'=0}^{n}\sum_{\tau=e}^{o}\left[A^{\sigma\ell n}_{\tau m'n'}\ \mathrm{Re}\vec{N}_{\tau m'n'}(\vec{r}-\vec{r}_i)\right. \tag{10b}$$

$$\left. + B^{\sigma\ell n}_{\tau m'n'}\ \mathrm{Re}\vec{M}_{\tau m'n'}(\vec{r}-\vec{r}_i)\right] ;$$

$$|\vec{r} - \vec{r}_i| < r_{ij} ,$$

where the geometry is shown in Fig. 2. Note that the minimum value of r_{ij} is 2a and $|\vec{r}-\vec{r}_i| < 2a$ for Eq. (6) to be valid so that the condition for use of Eq. (10) is always satisfied.

It then remains to expand the incident field $\vec{E}^0(\vec{r})$ in terms of an origin centered at the i-th scatterer:

$$\vec{E}^0(\vec{r}) = \hat{x}\ \exp(ik\zeta_i)\ \exp[ik\hat{z}\cdot(\vec{r}-\vec{r}_i)]$$

$$= e^{ik\zeta_i}\sum_{s=1}^{\infty}\left\{f_{o1s}\ \mathrm{Re}\vec{M}_{o1s}(\vec{r}-\vec{r}_i) + g_{e1s}\ \mathrm{Re}\vec{N}_{e1s}(\vec{r}-\vec{r}_i)\right\} \tag{11}$$

where $\zeta_i = \vec{r}_i\cdot\hat{z}$; f_{o1s}, g_{e1s} are the known expansion coefficients and $\hat{e} = \hat{x}$. Substituting Eqs. (10a), (10b) and (11) into Eq. (9) gives

$$\sum_{p=0}^{\infty}\sum_{m=0}^{p}\sum_{\tau=e}^{o}\left\{b^{i}_{\tau mp}\ \mathrm{Re}\vec{M}_{\tau mp} + c^{i}_{\tau mp}\ \mathrm{Re}\vec{N}_{\tau mp}\right\}$$

$$= e^{ik\zeta_i}\sum_{s=1}^{\infty}\left\{f_{o1s}\ \mathrm{Re}\vec{M}_{o1s} + g_{e1s}\ \mathrm{Re}\vec{N}_{e1s}\right\}$$

$$+ \sum_{j\neq i}^{N}\sum_{n=0}^{\infty}\sum_{\ell=0}^{n}\sum_{\sigma=e}^{o}\sum_{n'=0}^{\infty}\sum_{m'=0}^{n'}\sum_{\mu=e}^{o}\left\{(B^{j}_{\sigma\ell n}A^{\sigma\ell n}_{\mu m'n'} + C^{j}_{\sigma\ell n}B^{\sigma\ell n}_{\mu m'n'})\,\mathrm{Re}\vec{M}_{\mu m'n'}\right.$$

$$\left. + (B^{j}_{\sigma\ell n}B^{\sigma\ell n}_{\mu m'n'} + C^{j}_{\sigma\ell n}A^{\sigma\ell n}_{\mu m'n'})\,\mathrm{Re}\vec{N}_{\mu m'n'}\right\} . \tag{12}$$

Equation (12) is but an expansion of Eq. (3) in terms of vector spherical harmonics with respect to an origin O_i centered at the i-th scatterer. *A relation between the unknown expansion coefficients* $(b^i_{\tau mp}, c^i_{\tau mp})$ of the exciting field and the unknown expansion coefficients $(B^i_{\sigma\ell n}, C^i_{\sigma\ell n})$ of the scattered field can be obtained from Eq. (12) by using the known orthogonality properties of the vector spherical harmonics. It may be shown that

$$b^i_{o\ell n'} = e^{ik\zeta_i}\, f_{o\ell n'} + \sum_{j=1}^{N}{}' \sum_{n=0}^{\infty} \sum_{\ell=0}^{n} \sum_{\sigma=e}^{o} \left(B^j_{\sigma\ell n} A^{\sigma\ell n}_{o\ell n'} + C^j_{\sigma\ell n} B^{\sigma\ell n}_{o\ell n'}\right) \tag{13a}$$

$$c^i_{e\ell n'} = e^{ik\zeta_i}\, g_{e\ell n'} + \sum_{j=1}^{N}{}' \sum_{n=0}^{\infty} \sum_{\ell=0}^{n} \sum_{\sigma=e}^{o} \left(B^j_{\sigma\ell n} B^{\sigma\ell n}_{e\ell n'} + C^j_{\sigma\ell n} A^{\sigma\ell n}_{e\ell n'}\right) \tag{13b}$$

where $\sum'$ denotes the sum over all scatterers except the i-th scatterer.

Substituting Eq. (7) into the above expressions, we obtain a self-consistent set of equations for the unknown exciting field coefficients as given in Eqs. (14a,b).

$$\begin{aligned} b^i_{o\ell n'} = e^{ik\zeta_i}\, f_{o\ell n'} + \sum_{j=1}^{N}{}' \sum_{n=0}^{\infty} \sum_{\ell=0}^{n} \sum_{\sigma=e}^{o} \sum_{p=0}^{\infty} \sum_{m=0}^{p} \sum_{\mu=e}^{o} & \left[\left\{\left(T^{\sigma\ell n}_{\mu mp}\right)^{11} b^j_{\mu mp} + \left(T^{\sigma\ell n}_{\mu mp}\right)^{12} c^j_{\mu mp}\right\} A^{\sigma\ell n}_{o\ell n'}\right. \\ & \left. + \left\{\left(T^{\sigma\ell n}_{\mu mp}\right)^{21} b^j_{\mu mp} + \left(T^{\sigma\ell n}_{\mu mp}\right)^{22} c^j_{\mu mp}\right\} B^{\sigma\ell n}_{o\ell n'}\right] \end{aligned} \tag{14a}$$

$$\begin{aligned} c^i_{e\ell n'} = e^{ik\zeta_i}\, g_{e\ell n'} + \sum_{j=1}^{N}{}' \sum_{n=0}^{\infty} \sum_{\ell=0}^{n} \sum_{\sigma=e}^{o} \sum_{p=0}^{\infty} \sum_{m=0}^{p} \sum_{\mu=e}^{o} & \left[\left\{\left(T^{\sigma\ell n}_{\mu mp}\right)^{11} b^j_{\mu mp} + \left(T^{\sigma\ell n}_{\mu mp}\right)^{12} c^j_{\mu mp}\right\} B^{\sigma\ell n}_{e\ell n'}\right. \\ & \left. + \left\{\left(T^{\sigma\ell n}_{\mu mp}\right)^{21} b^j_{\mu mp} + \left(T^{\sigma\ell n}_{\mu mp}\right)^{22} c^j_{\mu mp}\right\} A^{\sigma\ell n}_{e\ell n'}\right] \end{aligned} \tag{14b}$$

Thus, the unknown scattered field expansion coefficients are eliminated through use of the T-matrix resulting in a set of equations involving the expansion coefficients (b_{mp}, c_{mp}) of the exciting field only. These coefficients are functions of the positions of all the scatterers.

3. CONFIGURATIONAL AVERAGING

In order to average the wave fields over the positions of all the scatterers we define the probability density function of finding the first scatterer at $\vec{r}_1$, the second scatterer at $\vec{r}_2$, and so forth by $p(\vec{r}_1, \vec{r}_2, \cdots, \vec{r}_N)$. This probability density function may be written as

$$\begin{aligned} p(\vec{r}_1, \vec{r}_2, \cdots, \vec{r}_N) &= p(\vec{r}_i)\, p(\vec{r}_1, \vec{r}_2, \cdots', \cdots, \vec{r}_N | \vec{r}_i) \\ &= p(\vec{r}_i)\, p(\vec{r}_j | \vec{r}_i)\, (p(\vec{r}_1, \vec{r}_2, \ldots', \ldots', \ldots \vec{r}_N | \vec{r}_i, \vec{r}_j) \\ &= \cdots \end{aligned} \tag{15}$$

where $p(\vec{r}_i)$ denotes the probability density of finding a scatterer at $\vec{r}_i$, while $p(\vec{r}_j | \vec{r}_i)$ denotes the conditional probability of finding a scatterer at $\vec{r}_j$ if a scatterer is known to be at $\vec{r}_i$. A prime in the first expansion of Eq. (15) means $\vec{r}_i$ is absent while two primes in the second expansion of Eq. (15) means both $\vec{r}_i$ and $\vec{r}_j$ are absent.

If the scatterers are randomly distributed, the positions of all scatterers are equally probable within the volume V accessible to the scatterers, and hence

$$p(\vec{r}_i) = \begin{cases} n_o / N & \vec{r}_i \in V \\ 0 & \vec{r}_i \notin V \end{cases} \tag{16}$$

where n_o is the uniform number density of the scatterers and N is the total number of scatterers. In addition, for nonoverlap of the imaginary spheres circumscribing each scatterer we approximate the conditional density as follows:

$$p(\vec{r}_j|\vec{r}_i) = \begin{cases} n_0/N & |\vec{r}_i-\vec{r}_j| \geq 2a \\ 0 & |\vec{r}_i-\vec{r}_j| < 2a \end{cases} \tag{17}$$

The form of the pair-correlation in Eq. (17) describes the usual radially symmetric distribution function with an exclusion surface or "hole" corresponding to a sphere of radius 2a. This form of the pair-correlation is expected to be satisfactory at low concentrations c where $c = n_0 \frac{4}{3}\pi a^3$ [18]. However, at higher concentrations Eq. (17) does not account for either the space occupying property of the scatterers nor the increase in local order with increasing c resulting from the well defined shape of the neighboring scatterers [2]. The available volume V_a for locating the center of the N-th scatterer randomly in the total volume V after the first N-1 scatterers have been previously located is a function of concentration. For hard spheres, the minimum value of $V_a = V - (N-1)\frac{4}{3}\pi(2a)^3 = V(1-8c)$. The maximum value is considerably higher than this. Since this restriction applies only to the centers of spheres and does not take into account their physical dimensions, this model is expected to be better in the Rayleigh regime when the spheres appear to be point particles (see Section 7). Thus, a realistic form of the pair correlation function must depend on concentration as well as the distance between the two scatterers under consideration. Twersky [2] has considered concentration dependent pair correlations and has extended the formalism to higher concentrations in the Rayleigh regime.

We denote the configurational average of a statistical quantity f as

$$\langle f\rangle_i = \int_V \cdots \int_V f\, p(\vec{r}_1,\vec{r}_2,\ldots|,\vec{r}_n|\vec{r}_i)\,d\vec{r}_1 d\vec{r}_2,\ldots|,d\vec{r}_n \tag{18a}$$

$$\langle f\rangle_{ij} = \int_V \cdots \int_V f\, p(\vec{r}_1,\vec{r}_2,\ldots|,\ldots|,\ldots\vec{r}_N|\vec{r}_i,\vec{r}_j)\,d\vec{r}_1 d\vec{r}_2,\ldots|,\ldots|,\ldots d\vec{r}_N \tag{18b}$$

where in Eq. (18a) we have averaged over all scatterers except the i-th and in Eq. (18b), over all scatterers except the i-th and j-th,

and so on. Multiplying both sides of Eqs. (14a,b) by the probability density given by Eq. (15) and using Eqs. (16)-(18), we obtain the configurational average of $b_{\tau mp}, c_{\tau mp}$

$$\langle b^i_{oln'} \rangle_i = e^{i\bar{k}\cdot\bar{r}_i} f_{oln'} + \frac{1}{V} \sum_{j=1}^{N,} \sum_{n,l,\sigma} \sum_{p,m,\tau} \int_{V'} \Big\{ \Big[\big(T^{\sigma l n}_{\tau mp}\big)^{11} \langle b^j_{\tau mp} \rangle_{ij}$$

$$+ \big(T^{\sigma l n}_{\tau mp}\big)^{12} \langle c^j_{\tau mp} \rangle_{ij} \Big] A^{\sigma l n}_{oln'} + \Big[\big(T^{\sigma l n}_{\tau mp}\big)^{21} \langle b^j_{\tau mp} \rangle_{ij}$$

$$+ \big(T^{\sigma l n}_{\tau mp}\big)^{22} \langle c^j_{\tau mp} \rangle_{ij} \Big] B^{\sigma l n}_{oln'} \Big\} \, d\bar{r}_j \qquad (19)$$

$$\langle c^i_{eln'} \rangle_i = e^{i\bar{k}\cdot\bar{r}_i} g_{eln'} + \frac{1}{V} \sum_{j=1}^{N,} \sum_{n,l,\sigma} \sum_{p,m,\tau} \int_{V'} \Big\{ \Big[\big(T^{\sigma l n}_{\tau mp}\big)^{11} \langle b^j_{\tau mp} \rangle_{ij}$$

$$+ \big(T^{\sigma l n}_{\tau mp}\big)^{12} \langle c^j_{\tau mp} \rangle_{ij} \Big] B^{\sigma l n}_{eln'} + \Big[\big(T^{\sigma l n}_{\tau mp}\big)^{21} \langle b^j_{\tau mp} \rangle_{ij}$$

$$+ \big(T^{\sigma l n}_{\tau mp}\big)^{22} \langle c^j_{\tau mp} \rangle_{ij} \Big] A^{\sigma l n}_{eln'} \Big\} \, d\bar{r}_j \qquad (20)$$

where V' denotes the volume of the medium excluding the volume of the hole of radius 2a. For identical scatterers, $\sum_{j=1}^{N,}$ can be replaced by (N-1). Equations (19,20) indicate that the conditional average with one scatterer fixed, viz., $[\langle b^i_{\tau mp} \rangle_i, \langle c^i_{\tau mp} \rangle_i]$ is given in terms of the conditional average with two scatterers fixed, viz., $[\langle b^j_{\tau mp} \rangle_{ij}, \langle c^j_{\tau mp} \rangle_{ij}]$. Lax [10] has suggested a quasi-crystalline approximation (QCA) to close the system:

$$\left.\begin{aligned} \langle b^j_{\tau mp} \rangle_{ij} &\simeq \langle b^j_{\tau mp} \rangle_j \\ \langle c^j_{\tau mp} \rangle_{ij} &\simeq \langle c^j_{\tau mp} \rangle_j \end{aligned}\right\} \; i \neq j \qquad (21)$$

The validity of this approximation is examined in Section 8.

4. THE COHERENT WAVE

The heirarchy of equations implied by Eqs. (19,20) is truncated by invoking the QCA, i.e., Eq. (21). A plane wave solution for the system of equations given by Eqs. (19) and (20) is assumed, using an effective wave number K to characterize the bulk medium:

$$
\begin{aligned}
b^i_{\ell mp}(\vec{r}_i) &= i^p Y_{\ell mp} e^{i\vec{K}\cdot\vec{r}_i} \\
c^i_{\ell mp}(\vec{r}_i) &= i^p Z_{\ell mp} e^{i\vec{K}\cdot\vec{r}_i}
\end{aligned}
\tag{22}
$$

where $Y_{\ell mp}$ and $Z_{\ell mp}$ are unknown constants. The effective wave number $\vec{K}$ is assumed to be parallel to that of the incident wave which in the present case is along the z-axis. Since the symmetry axes of the scatterers are also assumed parallel to the z-axis, the bulk medium is isotropic. Substitution of Eq. (22) in Eqs. (19) and (20) gives

$$
\begin{aligned}
i^{n'} Y_{oln'} e^{i\vec{K}\cdot\vec{r}_i} = e^{ikz_i} f_{oln'} &+ \frac{N-1}{V} \sum_{n,\ell,\sigma} \sum_{p,m,\ell} \delta_{\ell m}\, i^p \\
&\cdot \int_{V'} \left\{ \left[\left(T^{\sigma\ell n}_{\ell mp}\right)^{11} Y_{\ell mp} + \left(T^{\sigma\ell n}_{\ell mp}\right)^{12} Z_{\ell mp} \right] A^{\sigma\ell n}_{oln'} + \left[\left(T^{\sigma\ell n}_{\ell mp}\right)^{21} Y_{\ell mp} \right.\right. \\
&\left.\left. + \left(T^{\sigma\ell n}_{\ell mp}\right)^{22} Z_{\ell mp} \right] B^{\sigma\ell n}_{oln'} \right\} e^{i\vec{K}\cdot\vec{r}_j}\, d\vec{r}_j
\end{aligned}
\tag{23a}
$$

$$
\begin{aligned}
i^{n'} Z_{eln'} e^{i\vec{K}\cdot\vec{r}_i} = e^{ikz_i} g_{eln'} &+ \frac{N-1}{V} \sum_{n,\ell,\sigma} \sum_{p,m,\ell} \delta_{\ell m}\, i^p \\
&\cdot \int_{V'} \left\{ \left[\left(T^{\sigma\ell n}_{\ell mp}\right)^{11} Y_{\ell mp} + \left(T^{\sigma\ell n}_{\ell mp}\right)^{12} Z_{\ell mp} \right] B^{\sigma\ell n}_{eln'} + \left[\left(T^{\sigma\ell n}_{\ell mp}\right)^{21} Y_{\ell mp} \right.\right. \\
&\left.\left. + \left(T^{\sigma\ell n}_{\ell mp}\right)^{22} Z_{\ell mp} \right] A^{\sigma\ell n}_{eln'} \right\} e^{i\vec{K}\cdot\vec{r}_j}\, d\vec{r}_j
\end{aligned}
\tag{23b}
$$

where $\delta_{\ell n}$ is the Kronecker delta and applies to the T-matrix elements of rotationally symmetric scatterers, viz., no azimuthal mode coupling.

It remains to perform the integration over V' in Eqs. (23a,b), the details of which are given in Appendix A. To derive the dispersion relations, we apply the extinction theorem to the two sets of terms in Eq. (23) after integration (each satisfying the wave equation with wave numbers K and k) as discussed in detail by Twersky [4] and Varadan, Varadan and Pao [8]. According to the extinction theorem the integral I_∞ which is evaluated over a surface S_∞ precisely cancels the incident field (see Appendix A). By equating the remaining terms, we obtain an infinite system of equations for the unknowns:

$$i^{n'} Y_{oln'} = \frac{6c}{(ka)^2-(Ka)^2} \sum_{n=0}^{\infty} \sum_{p=0}^{\infty} \sum_{\lambda=|n-n'|}^{n+n'} i^p (-1)^\lambda (JH)_\lambda \left\{ Y_{olp} \left[\left(T^{oln}_{olp}\right)^{11} \psi_{oo}(n,n',\lambda) + \left(T^{eln}_{olp}\right)^{21} \chi_{eo}(n,n',\lambda) \right] + Z_{elp} \left[\left(T^{oln}_{elp}\right)^{12} \psi_{oo}(n,n',\lambda) + \left(T^{eln}_{elp}\right)^{22} \chi_{eo}(n,n',\lambda) \right] \right\} \tag{24a}$$

$$i^{n'} Z_{eln'} = \frac{6c}{(ka)^2-(Ka)^2} \sum_{n=0}^{\infty} \sum_{p=0}^{\infty} \sum_{\lambda=|n-n'|}^{n+n'} i^p (-1)^\lambda (JH)_\lambda \left\{ Y_{olp} \left[\left(T^{oln}_{olp}\right)^{11} \chi_{oe}(n,n',\lambda) + \left(T^{eln}_{olp}\right)^{21} \psi_{ee}(n,n',\lambda) \right] + Z_{elp} \left[\left(T^{oln}_{elp}\right)^{12} \chi_{oe}(n,n',\lambda) + \left(T^{eln}_{elp}\right)^{22} \psi_{ee}(n,n',\lambda) \right] \right\} . \tag{24b}$$

We define c as the effective "spherical" concentration which equals $4\pi a^3 n_o/3$. The term $(JH)_\lambda$ is given by

$$(JH)_\lambda = 2ka\ j_\lambda(2Ka)\ h'_\lambda(2Ka) - 2Ka\ h_\lambda(2ka)\ j'_\lambda(2Ka) . \tag{25}$$

The factors $\psi_{oo}(n,n',\lambda)$ and $\chi_{oe}(n,n',\lambda)$ are given in Appendix B. They are related to those given by Cruzan [17] which are also summarized in Appendix B.

The set of equations given in Eq. (24a,b) are homogeneous and linear in the unknowns (Y_{oln}, Z_{eln}). For a nontrivial solution, we require that the determinant of the coefficient matrix vanish which

yields a relation for the effective wave number K in terms of k and the T-matrix of the scatterer. This is the dispersion relation for the scatterer-filled medium. Equations (24a) and (24b) form a general expression valid for any arbitrary collection of scatterers provided the scatterers are identical and rotationally symmetric with parallel orientation along the $\bar{K}$ vector. Since the T-matrix is the only factor that contains information about the exact shape and boundary conditions at the scatterers, one can also use the above formalism for a collection of perfectly conducting, dielectric or multi-layered scatterers. The T-matrix for such various scatterers has been studied by many authors [23,24,25].

5. LOW FREQUENCY SOLUTIONS

In the Rayleigh or low-frequency limit, the size of the scatterer is considered to be very small compared to the incident wavelength. It is then sufficient to take only the lowest order coefficients in the expansion of the fields. In this limit, the elements of the T-matrix can be obtained in closed form for simple shapes such as sphere and spheroid [22]:

<u>Sphere</u>: $$\left(T^{ell}_{ell}\right)^{22} = \frac{2}{3}\, i(ka)^3 \left(\frac{\varepsilon_r - 1}{\varepsilon_r + 2}\right) + O(k^5a^5) \tag{26}$$

<u>Spheroid</u>: $$\left(T^{ell}_{ell}\right)^{22} \simeq \frac{i(ka)^3(\varepsilon_r - 1)f_2}{2(\varepsilon_r+2) - 3(\varepsilon_r-1)f_1(e)} \tag{27}$$

where e is defined by $e = \sqrt{(a/b)^2 - 1}$ for the oblate spheroid ('a' and 'b' being the semi-major and semi-minor axes, respectively). The functions f_1 and f_2 are given by

$$f_1(e) = e - \tan^{-1}e - \frac{1}{3} + \frac{1}{e^2} - \frac{\tan^{-1}}{e^3} \tag{28a}$$

$$f_2(e) = \frac{4}{3}\,\frac{1}{\sqrt{1+e^2}} \quad . \tag{28b}$$

From Eq. (24b) and using leading terms of the T-matrix of $O(k^3a^3)$, we obtain the following result for the unknown Z_{ell}:

$$Z_{ell} = \frac{6c}{(ka)^2 - (Ka)^2} \left[Z_{ell} \left(T^{ell}_{ell} \right)^{22} (JH)_0 + \frac{1}{2} \left(T^{ell}_{ell} \right)^{22} Z_{ell} (JH)_2 \right] \tag{29}$$

Dispersion relations are obtained by substituting Eq. (26) or (27) in Eq. (29) and using the leading term in the expansion for the Bessel and Hankel functions composing $(JH)_0$ and $(JH)_2$:

$$\underline{\text{Sphere}}: \quad \left(\frac{K}{k}\right)^2 = \frac{1 + 2c\left(\frac{\epsilon_r - 1}{\epsilon_r + 2}\right)}{1 - c\left(\frac{\epsilon_r - 1}{\epsilon_r + 2}\right)} \tag{30}$$

$$\underline{\text{Spheroid}}: \quad \left(\frac{K}{k}\right)^2 = \left\{ 1 + \frac{(3c/2)\,\frac{\epsilon_r - 1}{\epsilon_r + 2}\, f_2}{1 - \left(\frac{3}{2}\right)\left(\frac{\epsilon_r - 1}{\epsilon_r + 2}\right) f_1} \right\}$$

$$\times \left\{ 1 - \frac{\left(\frac{3c}{4}\right)\left(\frac{\epsilon_r - 1}{\epsilon_r + 2}\right) f_2}{1 - \frac{3}{2}\left(\frac{\epsilon_r - 1}{\epsilon_r + 2}\right) f_1} \right\}^{-1} . \tag{31}$$

Equation (30) is recognized as the dispersion relation of the Clausius-Mossotti form.

If the concentration $c \ll 1$, the dispersion relations simplify to

$$\underline{\text{Sphere}}: \quad \frac{K}{k} = 1 + \frac{3}{2} c\left(\frac{\epsilon_r - 1}{\epsilon_r + 2}\right) \tag{32}$$

$$\underline{\text{Spheroid}}: \quad \frac{K}{k} = 1 + \frac{(9c/8)\left(\frac{\epsilon_r - 1}{\epsilon_r + 2}\right) f_2}{1 - \frac{3}{2}\left(\frac{\epsilon_r - 1}{\epsilon_r + 2}\right) f_1} . \tag{33}$$

Equation (33) can be written in terms of the forward scattering amplitude $F(0)$:

$$\frac{\bar{K}}{K} = 1 + \frac{2\pi n_0}{k^2} F(0) \tag{34}$$

which in this case is valid only for very low concentrations and in the Rayleigh limit.

6. DISPERSION AT HIGHER FREQUENCIES

To study the dispersion at resonant and higher frequencies, we must consider higher powers of ka, and this implies that a large number of terms (Y_{oln}, Z_{eln}) must be kept in the expansion of the average field. This is best done numerically. A block diagram of the FORTRAN program written for this purpose is shown in Fig. 3. The blocks identify major subroutines which perform the following functions:

- **MAIN** The main program sets up the basic loops so as to calculate K for various frequencies (ka) and concentrations (c).
- **RDDATA** This subroutine is used to input data, e.g., scatterer size (ka), concentration (c), matrix sizes, etc.
- **RTINT** This subroutine calculates the initial guess for K in the Rayleigh limit at a given concentration c using Eqs. (30) or (31).
- **TMAT** This subroutine calculates the T-matrix for given scatterer shape and size (ka). Current maximum size is 40 × 40.
- **CGRTQ** This subroutine searches for the root in the complex plane (given an initial guess) by attempting to force the determinant of a coefficient matrix (C) = 0.
- **AUX** This subroutine sets up the coefficient matrix according to Eqs. (24a,b). Maximum size is 40 × 40.
- **AB** This subroutine calculates the factors α_{oo}, β_{oe} as given in Appendix B.
- **TRIXJ** This subroutine calculates the Wigner 3-j coefficients.
- **CXMTX** This subroutine calculates the det C for a given K using standard Gauss elimination.

The computational procedure is based on forming the coefficient matrix C according to Eqs. (24a,b). For a given ka, the roots of the equation $\det C = 0$ are searched in the complex K-plane using an iterative root searching algorithm which employs Muller's method. Good initial guesses for K were provided in the Rayleigh limit by Eqs. (30) or (31), and these could be used systematically

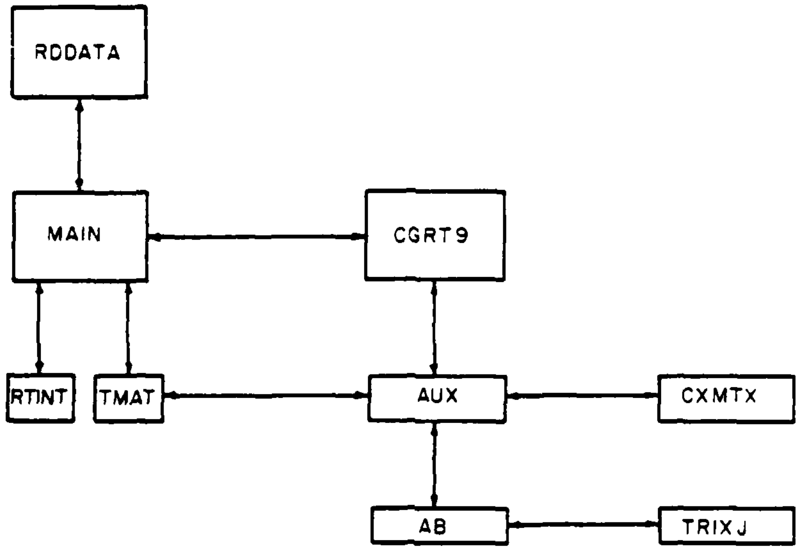

Figure 3. Block diagram of the computer program

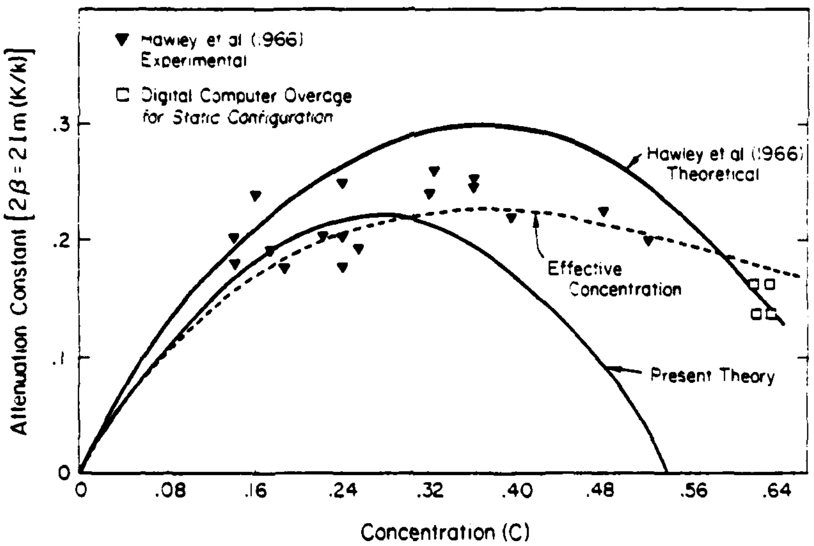

Figure 4. Attenuation vs concentration for a random distribution of dielectric spheres at ka = 11.8

to obtain convergence at increasingly higher values of ka. Similarly, the dependence of K on concentration at a fixed frequency could be computed. The real part of K, K_1, determines the phase velocity while the imaginary part, K_2 determines the coherent attenuation.

7. COMPUTATIONS AND COMPARISONS WITH EXPERIMENTS

A major aim of the computational method presented here is to provide a means of studying the dispersion characteristics of discrete random media at higher scatterer concentrations. Very few laboratory measurements of coherent wave attenuation and phase shift can be found in the open literature. However, results of two such experiments are presented here [13,14] and compared with computations.

The first experiment refers to the work of Hawley et al. [13] who measured the coherent wave attenuation and phase shift through a random assembly of dielectric spheres ($\epsilon_r = 1.034$) blown about by turbulence producing fans. The measurements were performed at fixed microwave frequency corresponding to $ka = 11.8$ and at various concentration levels up to maximum packing.. The attenuation measurements are shown in Fig. 4 together with computations using the present theory and that used by Hawley et al. The present theory agrees well with measurements up to $c = 0.32$, beyond which the attenuation is predicted to decrease and goes to zero at $c = 0.54$. The dashed curve is based on the concept of the "effective" concentration which accounts for the decreasing available volume as the number density increases. The "effective" concentration equals $c/(1-c)$ and the dashed curve in Fig. 4 reflects the attenuation constant as a function of this altered concentration. The computations now agree well with the measurements. The coherent phase shift, ϕ, relative to free space is plotted as a function of concentration c in Fig. 5 where the experimentally adjusted values and the computations are in good agreement. Attenuation in the Rayleigh limit ($ka = 0.05$) for the same scatterer properties is shown in Fig. 6 based on the present theory. The computations fail for $c > 0.125$ since Im(K) becomes negative. This feature is repeated in the Rayleigh limit for other values of ϵ_r as shown later. At very low concentrations, the form of the pair correlation function as given in Eq. (17) seems valid. As the concentration increases, the available volume for the other scatterers decreases.

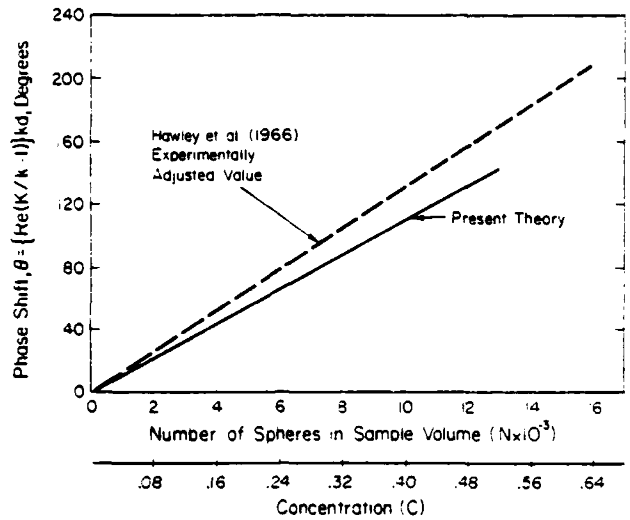

Figure 5. Phase shift vs concentration or number of spheres in sample volume for a random distribution of dielectric spheres at ka = 11.8

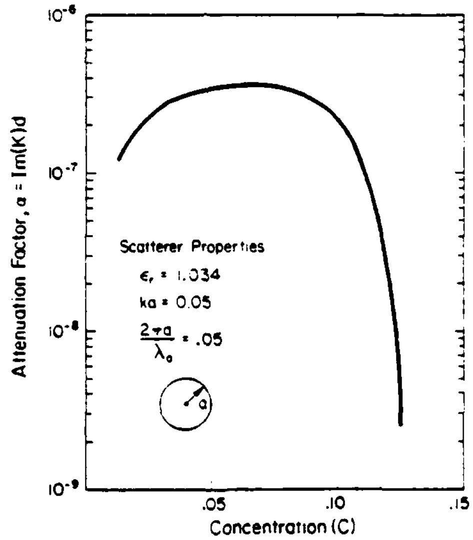

Figure 6. Attenuation factor vs concentration for a random distribution of dielectric spheres

Assuming that the centers of (N-1) scatterers are randomly located in a given volume V, the minimum available volume V_a for locating the center of the N-th scatterer is $V_a = V - \frac{4}{3}\pi(2a)^3(N-1) = V(1-8c)$ where it is assumed that each scatterer center is surrounded by a hole of radius $2a$ and that these holes do not interpenetrate. Now, $V_a \to 0$ as $c \to 0.125$ so this may explain why the computations cause $Im(K) \to 0$ for $c \to 0.125$, at least in the Rayleigh limit. Figure 7 shows computations in the Rayleigh limit for spheres with $\epsilon_r = 3.168$ and $ka = 0.05$ compared with the analytical results of Twersky [2] who obtained the leading effects of pair-correlation at low frequencies. His formula reduces to $Im(K) = \frac{1}{2} n_o \sigma_s W$ where $W = (1-c)^4/(1+2c)^2$, n_o is the number density and σ_s is the total scattering cross section of a lossless sphere. The agreement is good for $c < 0.05$ while great discrepancy is exhibited at higher concentrations. The concept of the "effective" concentration does not significantly alter the results at such low concentrations. To obtain better agreement at higher concentrations it is necessary to use a more realistic form for the pair-correlation function defined in Eq. (17). A factor containing some dependence of $p(\vec{r}_j|\vec{r}_i)$ on $\vec{r}_{ij}$ has to be introduced, but the form for this dependence constitutes a difficult problem in statistical geometry [18].

A second set of coherent wave measurements by Olsen and Kharadly [14] at relatively low concentrations ($c = 0.007, 0.014$) and at a single microwave frequency ($ka = 4.67$) was available for comparison with computations. Their measurement procedure, based on ensemble averaging made on a random collection of dielectric spheres ($\epsilon_r = 2.26$), exhibited greater control on the statistics of the scatterer distribution yielding accurate measurements with low standard errors. Attenuation as a function of ka is shown in Fig. 8 at $c = 0.014$. The experimental value is also shown with its estimated standard error. Figure 9 shows the same results at $c = 0.007$. As expected, the agreement between theory and experiment is excellent at these low concentrations. Figures 10 and 11 show the phase shift relative to free space as a function of ka for $c = 0.014$ and 0.007, respectively, together with the experimental values. Again agreement is good and within the estimated standard errors. Figure 12 shows the computation of attenuation vs. concentration at $ka = 4.67$. The values based on single scattering theory, see

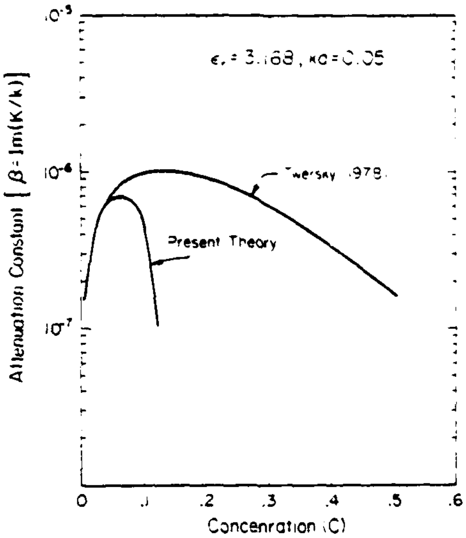

Figure 7. Attenuation factor vs concentration for a random distribution of dielectric spheres

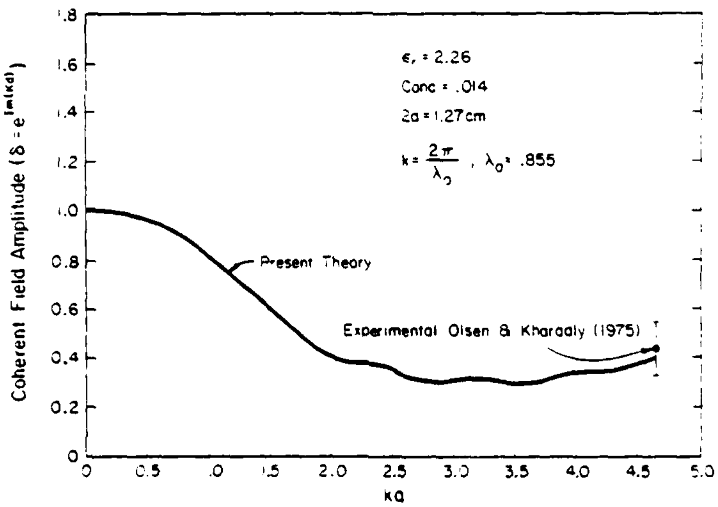

Figure 8. Coherent field amplitude vs ka for a random distribution of dielectric spheres; comparison with experimental value of Olsen and Kharadly (1975) at ka = 4.67

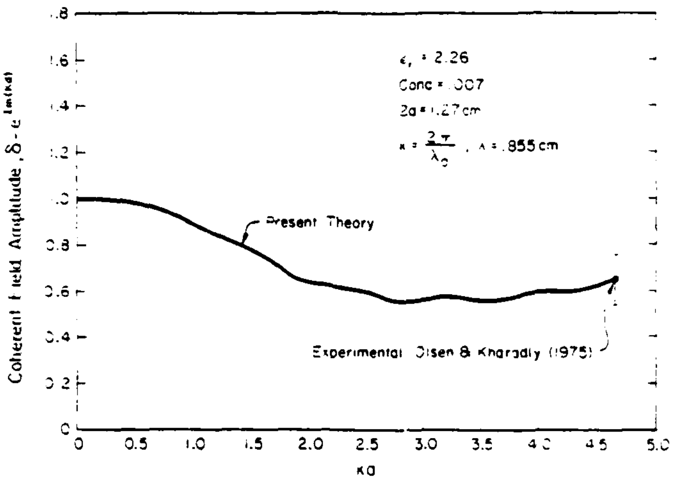

Figure 9. Coherent field amplitude vs ka for a random distribution of dielectric spheres; comparison with experimental value of Olsen and Kharadly (1975) at ka = 4.67

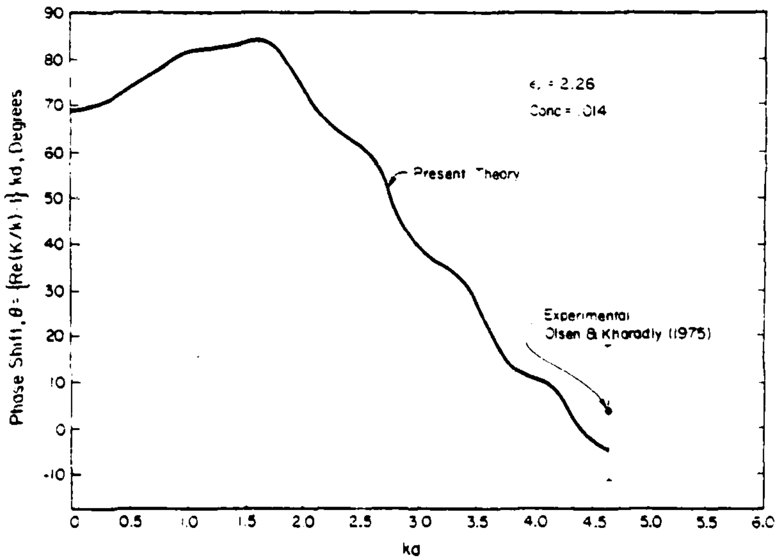

Figure 10. Phase shift vs ka for a random distribution of dielectric spheres; comparison with experimental value of Olsen and Kharadly (1975) at ka = 4.67

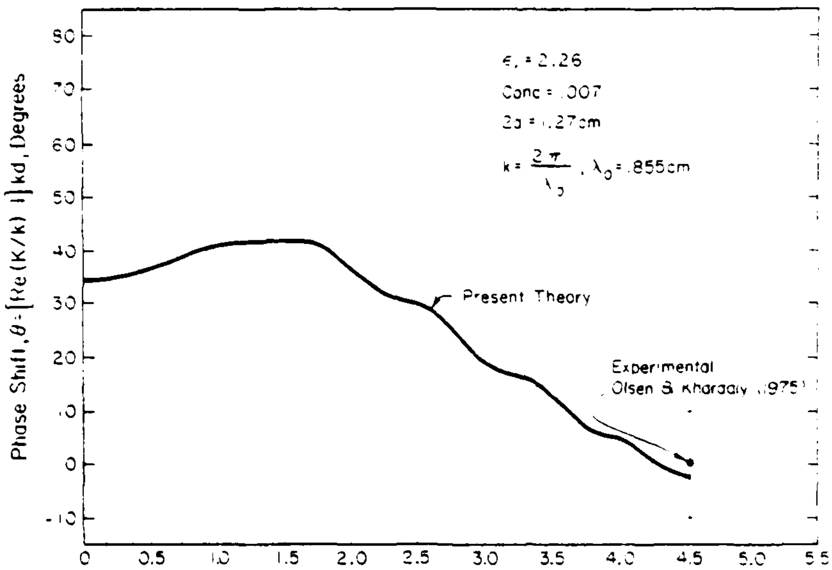

Figure 11. Phase shift vs ka for a random distribution of dielectric spheres: comparison with experimental value of Olsen and Kharadly (1975) at ka = 4.67

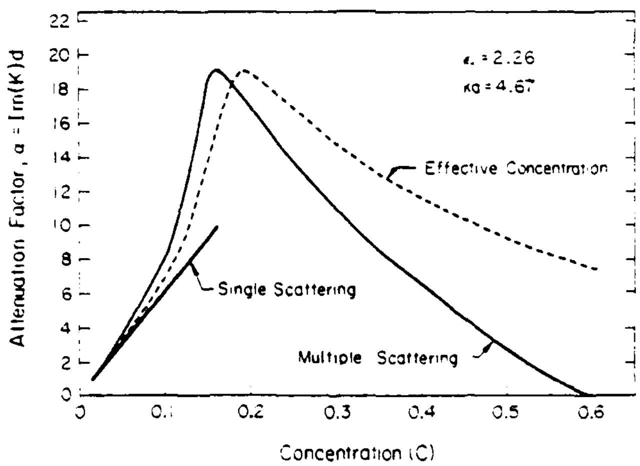

Figure 12. Attenuation vs concentration for a random distribution of dielectric spheres

Eq. (34), are also shown together with the dashed curve based on the "effective" concentration concept. In this case, single scattering theory seems to be valid for $c < 0.05$. The computations cause $Im(K) < 0$ for $c \geq 0.6$ while the dashed curve does not exhibit this phenomenon. Again, the validity of the computations at higher concentrations is not established. Previous computations presented in [6,7] for concentrations of 0.20, as a function of ka (or frequency), exhibit certain "null" characteristics for the attenuation at certain ka values. This phenomenon is not real and is caused by $Im(K) < 0$ at these values. However, the computations presented in [6,7] for $c = 0.05$ and 0.10 are correct.

8. QUASI-CRYSTALLINE APPROXIMATION (QCA)

It is instructive to examine the physical implications of terms of the form $\langle b^j_{mp} \rangle_{ij}$ that occur in Eq. (21). This is the exciting field coefficient of the j-th scatterer when the positions of all scatterers except the j-th and any other scatterer denoted by 'i' are averaged over. When the number of particles in the system is large this may be thought of as the field exciting the i-th scatterer in an effective, macroscopically homogeneous medium containing two scatterers 'i' and 'j'. The QCA implicitly omits all multiple scattering processes that can take place between 'i' and 'j'. Efforts have been made to restore such scattering processes by Twersky [2] and by Schwartz and Ehrenreich [19] who, in a different context, discuss the contribution of clusters of two or more particles as in systems with short range order.

In the classical context, it is also important to discuss the dependence of QCA on the frequencies under consideration as well as on the concentration of the scatterers. Scattering in a two scatterer configuration is quite dependent on the ratio of the distance '2d' between the scatterers to the overall dimensions '2a' of the scatterer. Numerical computations suggest that multiple scattering effects at any frequency of the incident wave will be small if d/a is large. Thus, the QCA is expected to be good for sparse concentrations, < 1%. At such concentrations the pair correlation function as used in Eq. 17 is also expected to be good. The analytical results we obtain at long wavelengths give ample proof of this as shown in the previous sections. Most authors cited previously as well as Talbot and Willis [20] in their recent work seem to agree

that the QCA is good at sparse concentrations. However, most of these papers present only long wavelength results. Further, Lax [21] comments immediately following the definition of the QCA that it is good for dense systems and exactly valid for systems with a crystalline structure. These are contradictory observations about the same approximation and needs to be studied further.

Our computations for various concentrations, but more importantly for various values of ka (the non-dimensional wavenumber) suggest that even at low values of $ka \sim 0.05$, the model fails if the volume concentration c exceeds 12.5%, whereas at ka values ~ 2.0 or more, the QCA leads to reasonable results for the bulk propagation constant at all values of the concentration in spite of the poor model used for the pair correlation function. We expect all types of multiple scattering effects including cluster effects to be important at concentrations ~ 0.1 or more, but it should be noted that the QCA type approximation neglects only repeated scattering between pairs or within a group of scatterers. It is also important to note that at wavelengths comparable to obstacle size and higher, i.e., $ka > 3.0$, the scattering is mostly in the forward direction. Thus, in this case repeated scattering should not be important, since the backscattered wave is significantly smaller than the forward scattered wave. This would help satisfy the QCA and may explain why the computations shown in Fig. 4 for $ka = 11.8$ are in reasonable agreement with experiment at all values of the effective concentration. It would appear that in the context of classical systems, Lax's statement about the validity of the QCA for dense systems should be qualified by the phrase, 'at high frequencies.' It would be interesting to study how the results will change with improved models of the pair correlation function. Then it would be possible to comment on the *sensitivity* of the results to the effect of the pair correlation function and the QCA separately.

9. RECOMMENDATIONS FOR FUTURE WORK

It is obvious from the preceding discussions on the QCA as well as the numerical results that the two major improvements required are for the QCA as well as the pair correlation function, so that good results can be obtained for all concentrations even at long and intermediate values of the wavelength. In a review article, Lax [21] has suggested that in the quantum mechanical context, the QCA

could be improved by using modified propagators for the fields. In the classical context, this implies that on the average, single particle scattering takes place in a macroscopically homogeneous medium, and, in this respect, this idea is the same as the coherent potential approximation (CPA) of Solid State Physics. The repeated multiple scattering between pairs of scatterers or cluster effects can be improved by making the self consistent approximation (SCA) in addition to the CPA.

For the purpose of discussion of these ideas within the T-matrix formalism given earlier, we denote by $\bar{E}^s_j$ and $\bar{E}^e_j$ the fields scattered by and exciting the j-th scatterer, respectively. The expansion coefficients of these fields as given in Eqs. (5,6) are denoted by B^j and b^j, respectively, omitting all subscripts.

The CPA can be expressed succinctly as

$$\langle B^j \rangle_j = T(K) \langle b^j \rangle_j \tag{35}$$

where the T-matrix relating the exciting and scattered field coefficients is evaluated using the bulk propagation constant K for the embedding medium. Thus the CPA implies that the field scattered by a single obstacle in the presence of several others when averaged over the position of all scatterers is the same as the field that would be produced by a single particle embedded in a macroscopically homogeneous medium described by the propagation constant K. The incorporation of the CPA into the previous formalism involves changes only in the computations and a redefinition of the T-matrix. It would be interesting to see the change, if any, in the numerical computations as a result of invoking the CPA.

The idea behind the 'self consistent approximation' (SCA) is somewhat more subtle. From the discussion in the section on the QCA, it is now clear that QCA-CPA neglects multiple scattering between two fixed scatterers. The SCA as defined by Schwartz and Ehrenreich [19] restores this by stating that

$$\langle B^j \rangle_{ij} = \tilde{T}(K) \langle b^j \rangle_j \tag{36}$$

where $\tilde{T}(K)$ is the 'T-matrix' of scatterer 'j' in the presence of scatterer 'i', in the effective medium with propagation constant K. Expressions for $\tilde{T}(K)$ as given by Peterson and Ström [16] may be written as

$$\tilde{T}(K) = R(\vec{r}_{ij}/2)\,T[1 - \sigma(-\vec{r}_{ij})T\sigma(\vec{r}_{ij})T]^{-1}$$

$$[1 + \sigma(-\vec{r}_{ij})TR(\vec{r}_{ij})]\; R(-\vec{r}_{ij}/2) \qquad (37)$$

where σ is a compact notation for the translation matrices A and B introduced in Eq. (10). The R matrix is simply the part of σ that is regular at the origin, i.e., for $\vec{r}_{ij} = 0$. All matrices in Eq. (37) are obtained using the bulk propagation constant for the host medium.

We observe that $\tilde{T}(K)$ explicitly depends on r_{ij} the distance between 'i' and 'j' and hence will be involved in the integrations in Eqs. (A1-A12). The integration procedure will no longer be simple as before and the SCA may be rather difficult to enforce in computations, especially if more realistic models are chosen for the pair correlation function.

Improvements to the pair correlation function must take into account the increase in short range order with increasing concentration in addition to enforcing the condition of no interpenetration of particles. Talbot and Willis [20] in their recent work have reviewed several models that depend on concentration. They discuss in particular two models by Matern [26] that depend on the concept of an available volume and hence are valid only for $c < 12.5\%$. Talbot and Willis also suggest the Percus-Yevick [27] model for the pair correlation function which is valid for $c < 0.3$.

Incorporation of the CPA as well as improved models of the pair correlation function into our computations are in progress. We hope that they will shed some light on the sensitivity of multiple scattering theories to approximations like QCA and SCA as a function of frequency and scatterer concentration. Needless to say additional experimental results are required for comparison with these computations.

APPENDIX A

Consider the following integral which appears in Eq. (23a):

$$\int Y_{lp}\, A_{oln}^{\ell n}\, e^{i\vec{K}\cdot\vec{r}_j}\, d\vec{r}_j \quad .$$

We observe that $A^{\cdot n}_{oln'}$ contains a term $\frac{\sin}{\cos}(\ell-1)\phi_{ij}$ which upon integration vanishes for $\ell \neq 1$. Also, only certain combinations of ℓ, p yield a non-zero value upon integration in conjunction with the properties of the T-matrix elements for rotationally symmetric scatterers. Thus the above integral reduces to evaluation of

$$\int_{V'} Y_{olp} A^{oln}_{oln'} e^{i\vec{K}\cdot\vec{r}_j} d\vec{r}_j = \sum_{\lambda = |n-n'|}^{n+n'} \gamma_{oo}(n,n',\lambda) Y_{olp} \int_{|\vec{r}_i - \vec{r}_j| > 2a} \cdot h_\lambda(kr_{ij}) P_\lambda(\cos\theta_{ij}) e^{i\vec{K}\cdot\vec{r}_j} d\vec{r}_j \qquad (A1)$$

where the expansion for $A^{oln}_{oln'}$ has been used and $\gamma_{oo}(n,n',\lambda)$ contains certain combination of Wigner 3-j symbols [16]. Now, we consider the integral in (A1)

$$I_A = \int_{|\vec{r}_i - \vec{r}_j| > 2a} e^{i\vec{K}\cdot\vec{r}_j} h_\lambda(kr_{ij}) P_\lambda(\cos\theta_{ij}) d\vec{r}_j \; . \qquad (A2)$$

Since $\vec{K} = K\hat{z}$ and $\vec{r}_j = \vec{r}_i - \vec{r}_{ij}$, we expand $\exp(-iK\hat{z}\cdot\vec{r}_{ij})$ in terms of the spherical harmonics

$$\exp(-iK\hat{z}\cdot\vec{r}_{ij}) = \exp(-iKr_{ij}\cos\theta_{ij}) = \sum_{n=0}^{\infty} (-1)^n i^n (2n+1) j_n(Kr_{ij}) \cdot P_n(\cos\theta_{ij}) \qquad (A3)$$

Substituting (A3) in (A2), we obtain

$$I = \int_{r_{ij} > 2a} j_n(Kr_{ij}) P_n(\cos\theta_{ij}) h_\lambda(kr_{ij}) P_\lambda(\cos\theta_{ij}) d\vec{r}_j \qquad (A4)$$

Observe that the two terms $j_n(Kr_{ij})P_n$ and $h_\lambda(kr_{ij})P_\lambda$ satisfy the scalar wave equation with wave numbers K and k, respectively. Defining

$$\alpha_\ell = h_\ell(kr_{ij})P_\ell(\cos\theta_{ij})$$

$$\beta_n = j_n(Kr_{ij})P_n(\cos\theta_{ij})$$

and using $\nabla^2\alpha_\ell = -k^2\alpha_\ell$ and $\nabla^2\beta_n = -K^2\beta_n$ in (A4), we obtain

$$I = \int_{r_{ij}>2a} \frac{1}{k^2-K^2}\left[\alpha_\ell\nabla^2(\beta_n) - \beta_n\nabla^2(\alpha_\ell)\right] d\bar{r}_j \;. \tag{A5}$$

Use of Green's theorem in (A5) reduces it to

$$I = \oint_{S_\infty,S_{2a}} \frac{1}{k^2-K^2}\left[\alpha_\ell\nabla(\beta_n) - \beta_n\nabla(\alpha_\ell)\right]\cdot d\bar{S} \tag{A6}$$

where S_∞ refers to the surface of a sphere of large radius centered around the j-th scatterer and S_{2a} refers to the surface of the hole centered around the j-th scatterer. The integral over S_{2a} can be evaluated in closed form

$$\oint_{S_{2a}} \frac{1}{k^2-K^2}\left[\alpha_\ell\nabla(\beta_n) - \beta_n\nabla(\alpha_\ell)\right]\cdot(-\hat{n})(2a)^2\sin\theta_{ij}\, d\theta_{ij}\, d\phi_{ij}$$

where $\hat{n}$ is the unit outward normal to S_{2a}. The above integral reduces to

$$\frac{(2a)^2}{k^2-K^2}\oint_{S_{2a}}\left(-\alpha_\ell\frac{\partial\beta_n}{\partial n} + \beta_n\frac{\partial\alpha_\ell}{\partial n}\right)\sin\theta_{ij}\, d\theta_{ij}\, d\phi_{ij} \tag{A7}$$

Upon substituting the previously defined expressions for α_ℓ and β_n and using the orthogonality of the Legendre polynomials in (A7), we get

$$\frac{8\pi a^2}{k^2-K^2}(JH)^*_\ell\,\frac{1}{(2\ell+1)}\,\delta_{\ell n}\;;\quad (JH)^*_\ell = (JH)_\ell\; a \tag{A8}$$

where $(JH)_\ell$ is given by Eq. (25) and $\delta_{\ell n}$ is the Kornecker delta. Hence, I defined in (A6) reduces to

$$I = I_\infty' + \frac{8\pi a^2}{k^2 - K^2} (JH)^* \frac{1}{2n+1} \; n \qquad \text{(A9)}$$

where I_∞' refers to the integration over S_∞ which is obtained by using the asymptotic expansion for $h_n(kr_{ij})$ as $r_{ij} \to \infty$ on S_∞. The integral I_A defined in (A2) now reduces to

$$I_A = I_\infty + \frac{8\pi a^2}{k^2 - K^2} (JH)^* e^{i\vec{K}\cdot\vec{r}_i} \ldots \qquad \text{(A10)}$$

Finally, the integral given in (A1) reduces to

$$Y_{oip} A_{oin'}^{oln} e^{i\vec{K}\cdot\vec{r}_i} d\vec{r}_i = \sum_{\ell = n-n'}^{n+n'} \ldots_{oo}(n,n',\ell) \ldots I_\infty + \frac{8\pi a^2}{k^2 - K^2} \ldots$$

$$\cdot (JH)^* e^{i\vec{K}\cdot\vec{r}_i} \qquad \text{(A11)}$$

In a similar manner, the remaining integrals in Eq. (23a,b) can be evaluated.

APPENDIX B

The translational operators (n,n',ℓ) and (n,n',ℓ) are given in (16):

$$\ldots_{oo}(n,n',\ell) = \ldots_{ee}(n,n',\ell)$$

$$= (-i)^{n'-n+\ell} \left[\frac{(2\ell+1)(2n'+1)}{2n'(n'+1)}\right] \left[\frac{n(n+1)}{n'(n'+1)}\right]^{1/2}$$

$$\cdot [n(n+1) + n'(n'+1) - \ell(\ell+1)] \begin{pmatrix} n & n' & \ell \\ \ldots & \ldots & \ldots \end{pmatrix} \begin{pmatrix} n & n' & \ell \\ \ldots & -1 & \ldots \end{pmatrix} \qquad \text{(B1)}$$

where $\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix}$ is the Wigner 3-j symbol (17).

$$\tau_{eo}(n,n',\nu) = -\tau_{oe}(n,n',\nu)$$

$$= i(-1)^{n'-n+\nu}\left[\frac{(2\nu+1)(2n'+1)}{2n'(n'+1)}\right]\left[\frac{n(n+1)}{n'(n'+1)}\right]^{-1/2}$$

$$\cdot\left[\nu^2 - (n-n')^2\right]\cdot\left[(n+n'+1)^2-\nu^2\right]^{-1/2}$$

$$\cdot\begin{pmatrix} n & n' & \nu-1 \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} n & n' & \nu \\ 1 & -1 & 0 \end{pmatrix} \tag{B2}$$

The above factors can be expressed in terms of factors given by Cruzan [17] as follows

$$\tau_{oo}(n,n',\nu) = \tau_{ee}(n,n',\nu) = (-1)^{\nu}\, a(n,n',\nu)\; a(1,n,-1,n'\,|\,\nu) \tag{B3}$$

$$\tau_{eo}(n,n',\nu) = -\tau_{oe}(n,n',\nu) = (-i)\, b(n,n',\nu)\; a(1,n,-1,n'\,|\,\nu,\nu-1) \tag{B4}$$

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Coherent Wave Attenuation by a Random Distribution of Particles †

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ABSTRACT

The coherent electromagnetic wave attenuation in an infinite medium composed of a random distribution of identical, finite scatterers is studied. A self-consistent multiple scattering theory using the T-matrix of a single scatterer and a suitable averaging technique is employed. The statistical nature of the position of scatterers is accounted for by ensemble averaging. This results in a hierarchy of equations relating the different orders of correlations between the scatterers. Lax's quasicrystalline approximation (QCA) is used to truncate the hierarchy enabling passage to a homogeneous continuum whose bulk propagation characteristics such as phase velocity and coherent wave attenuation can then be studied. Three models for the pair correlation function are considered. The Matern model and the well stirred approximation (WSA) are good only for sparse concentrations, while the Percus-Yevick approximation (P-YA) is good for a wider range of concentration. The results obtained using these models are compared with the available experimental results for dielectric scatterers embedded in another dielectric medium. Practical applications of this study include radar meterology and communications through hydrometers, dust, vegetation, etc.

1. INTRODUCTION

We consider the propagation of plane coherent electromagnetic waves in an infinite medium containing identical, lossless randomly distributed particles. Our aim is to characterize the random medium by an effective complex wave number K which would be a function of the particle concentration, electrical size and the statistical description of the random positions of the scatterers. The imaginary part of K describes the coherent attenuation which is due to multiple scattering only since the particles themselves are assumed to be lossless. The understanding of the behavior of Im(K) as a function of particle concentration c and/or frequency ka is very important in many practical applications, including wave propagation in the atmosphere and oceans and whenever distribution of random scatterers influence electromagnetic wave behavior.

The theoretical formulation presented here closely follows the procedure described in Varadan et. al. [1979] and Bringi et. al. [1981]. This approach is based on a self-consistent multiple scattering theory and relies on the T-matrix [Waterman 1971] which relates the field scattered by a particle to an arbitrary exciting field. The statistical description of the random position of the scatterers is used to define a configurational average which results in a hierarchy of equations relating the different orders of correlations between the scatterers. Lax's [1952] quasi-crystalline approximation is used to truncate the hierarchy which results in the usual "hole-correction" integrals. Following Twersky [1977, 1978 a,b], a radially symmetric pair-correlation function is introduced and approximate models are chosen from Talbot and Willis [1980].

The "well-stirred" approximation (WSA) was used previously by Varadan et. al. [1979] and Bringi et. al. [1981] which assumes no correlation between the particles except that they should not inter-penetrate. In particular, the WSA gives unphysical results for $c \geq 0.125$ at the Raleigh or low frequency limit.

In this paper, we consider two other pair-correlation functions, viz. (i) the Matern [1960] model and (ii) the Percus-Yevick [1957] model for a classical system of hard spheres. Computations of Im(K) are presented for dielectric scatterers in a dielectric medium, using the above three models as a function of frequency and concentration. We also compare our solution to some recent optical propagation experiments conducted by Ishimaru [1981]. Sample computations are also presented comparing the WSA and the single scattering approximation for a rain medium.

2. FORMULATION OF THE PROBLEM

Consider N identical, finite dielectric scatterers that are randomly distributed either in free space or in a different dielectric medium. The scatterers are homogeneous with a relative dielectric constant of ε_r, their centers being denoted by 0_1, 0_2, 0_3, ..., 0_N. They are assumed to be bodies of revolution with symmetry axis parallel to the z-direction. Monochromatic plane coherent electromagntic wave is assumed to propagate along the symmetry axis of the scatters to satisfy the condition that the effective medium be isotropic and polarization insensitive. The time dependence of the incident field and hence the fields scattered by the individual scatterers is all of the form $\exp(-j\omega t)$ and this is suppressed in the equations that follow.

Even though the theory presented here is valid for spheroidal scatterers [Varadan, et. al. 1981], we present numerical results only for spherical scatterers in order to compare our results with available experiments.

Let $\vec{E}^0(\vec{r})$ be the electric field arising from the incident plane wave and $\vec{E}_i^S(\vec{r})$ the field scattered by the i-th scatterer. Both these fields satisfy the vector Helmholtz equation. The problem at hand reduces to computing the total wave field at any point outside the scatterers, satisfying the appropriate boundary condition on the surface of the scatterers and radiation conditions at infinity.

The total field at any point outside the scatterers can be interpreted as the sum of the incident field and the fields scattered by all the scatterers, which can be written as

$$\vec{E}(\vec{r}) = \vec{E}^0(\vec{r}) + \sum_{i=1}^{N} \vec{E}_i^S(\vec{\rho}_i) \quad ; \quad \vec{\rho}_i = \vec{r}-\vec{r}_i \tag{1}$$

where $\vec{E}_i^S(\vec{\rho}_i)$ is the field scattered by the i-th scatterer at the observation point $\vec{r}$. However, the field that excites the i-th scatterer is the incident field $\vec{E}^0$ plus the fields scattered from all other scatterers except the i-th. The term exciting field $\vec{E}^e$ is used to distinguish between the field actually incident on a scatterer and the external incident field $\vec{E}^0$ produced by a source at infinity. Thus, at a point $\vec{r}$ in the vicinity of the i-th scatterer, we write

$$\vec{E}_i^e(\vec{r}) = \vec{E}^0(\vec{r}) + \sum_{j\neq i}^{N} \vec{E}_j^S(\rho_j) \quad ; \quad a \leq |\rho_j| < 2a \tag{2}$$

where 'a' is a typical dimension of the scatterer.

The exciting and scattered fields for each scatterer can be expanded in terms of vector spherical functions with respect to an origin at the center of that scatterer:

$$\vec{E}_i^e(\vec{r}) = \sum_{\tau=1}^{2} \sum_{\ell=1}^{\infty} \sum_{n=0}^{\ell} \sum_{\sigma=1}^{2} b^i_{\tau\ell n\sigma} \operatorname{Re} \vec{\psi}_{\tau\ell n\sigma}(\vec{r}_i)$$

$$= \sum_{\tau n} b^i_{\tau n} \operatorname{Re} \vec{\psi}^i_{\tau n} \tag{3}$$

$$\vec{E}_i^S(\vec{r}) = \sum_{\tau n} B^i_{\tau n} \vec{\psi}^i_{\tau n} \tag{4}$$

where the vector spherical functions are defined as

$$\vec{\psi}_{1\ell n\sigma}(\vec{r}) = \nabla \times [\vec{r}\, h_\ell^{(1)}(kr)]\, Y_{\ell n\sigma}(\theta,\phi) \tag{5}$$

$$\vec{\psi}_{2\ell n\sigma}(\vec{r}) = \frac{1}{k} \nabla \times \vec{\psi}_{1\ell n\sigma}(\vec{r}) \ . \tag{6}$$

In equations (3-6), k is the wave number; $h_\ell^{(1)}$ is the Hankel function of the first kind and the $Y_{\ell n\sigma}(\theta,\phi)$ are the normalized spherical harmonics defined with real angular functions. In Equation(3), the exciting field is expanded in terms of the regular (Re) basis set (Re $\vec{\psi}^i_{\tau n}$) obtained by replacing $h_n^{(1)}$ in Equations (5-6) by j_n, the spherical Bessel functions of the first kind. Thus, the choice of the basis set in Equation (4) satisfies the radiation condition at infinity for the scattered field, while the choice in (3) satisfies the

regular behavior of the exciting field in the region $a < |\vec{\rho}_i| < 2a$. The superscript i on the basis functions refer to expansions with respect to 0_i, and $b^i_{\tau n}$ and $B^i_{\tau n}$ are the unknown exciting and scattered field coefficients. We also expand the incident field in terms of vector spherical functions:

$$\vec{E}^0(\vec{r}) = e^{ik\hat{z}\cdot\vec{r}_i} \sum_{\tau n} a_{\tau n} \operatorname{Re} \vec{\psi}^i_{\tau n}(\vec{\rho}_i) \tag{7}$$

where $a_{\tau n}$ are the known incident field coefficients.

The unknown coefficients $b^i_{\tau n}$ can be related to $B^i_{\tau n}$ by means of any convenient scattering operator, in this case we employ the T-matrix as defined by Waterman [1971]:

$$B^i_{\tau n} = \sum_{\tau' n'} T^i_{\tau n,\tau' n'} \, b^i_{\tau' n'} \, . \tag{8}$$

Substituting Equations (3), (4) and (7) in (2), we obtain

$$\sum_{\tau n} b^i_{\tau n} \operatorname{Re} \vec{\psi}^i_{\tau n} = e^{ik\hat{z}\cdot r_i} \sum_{\tau n} a_{\tau n} \operatorname{Re} \vec{\psi}^i_{\tau n} + \sum_{j\neq i}^{N} \sum_{\tau n} B^j_{\tau n} \vec{\psi}^j_{\tau n} \tag{9}$$

Since the field quantities are expanded with respect to centers of each scatterer, we obtain Equation (9) with basis functions expanded with respect to i-th and j-th centers. In order to express them with respect to a common origin 0_i, we employ the translation and addition theorems for the vector spherical functions [see, for example, Boström, 1980] which may be written in a compact form as follows

$$\vec{\psi}_{\tau n}(\vec{\rho}_j) = \begin{cases} \sum_{\tau' n'} \sigma_{\tau n,\tau' n'}(\vec{\rho}_{ij}) \, Re\, \vec{\psi}_{\tau' n'}(\vec{\rho}_i) & ; \ |\vec{\rho}_{ij}| > |\vec{\rho}_i| \\ \\ \sum_{\tau' n'} R_{\tau n,\tau' n'}(\vec{\rho}_{ij}) \, \vec{\psi}_{\tau' n'}(\vec{\rho}_i) & ; \ |\vec{\rho}_{ij}| < |\vec{\rho}_i| \end{cases} \tag{10}$$

where $\vec{\rho}_{ij} = \vec{r}_i - \vec{r}_j$ is the vector connecting 0_j to 0_i, $\sigma_{\tau n,\tau' n'}$ is the translation matrix for the vector functions and $R_{\tau n,\tau' n'}$ is a matrix with spherical Hankel functions in $\sigma_{\tau n'\tau' n'}$ replaced by spherical Bessel functions.

Employing Equations (8) and (10) in (9) and using the orthogonality of the vector spherical basis functions, we obtain the following set of coupled algebraic equations for the exciting field coefficients $b^i_{\tau n}$:

$$b^i_{\tau n} = e^{ik\hat{z}\cdot\vec{r}_i} a_{\tau n} + \sum_{j\neq 1}^{N} \sum_{\tau n} \sum_{\tau' n'} \sigma_{\tau n,\tau' n'}(\vec{\rho}_{ij}) \, T^j_{\tau n,\tau' n'} \, b^j_{\tau' n'} \tag{11}$$

From Equation (11), it can be seen that the exciting field coefficients of the i-th scatterer explicitly depend on the position and orientation of the other scatterers. In this paper, we consider a random distribution of spherical scatterers and the case when $N \to \infty$ and the volume occupied by the scatterers $V \to \infty$ such that $N/V = n_o$ is a finite number density. For such distribution, a configurational average of Equation (11) can be made over the positions of all scatterers [see Varadan et. al., 1981] with QCA [Lax, 1952] to arrive at an equation for the configurational average $\langle b^i_{\tau n}\rangle_i$ of the exciting field coefficients with one scatterer fixed:

$$\langle b^i_{\tau n}\rangle_i = e^{ik\hat{z}\cdot\vec{r}_i} a_{\tau n} + (N-1) \sum_{\tau n'} \sum_{\tau'' n''} T_{\tau' n',\tau'' n''} \int_V p(\vec{r}_j|\vec{r}_i) \, \sigma_{\tau' n',\tau'' n''}(\vec{\rho}_{ij}) \, \langle b^j_{\tau n}\rangle_j \, d\vec{r}_j \tag{12}$$

where $p(\vec{r}_j|\vec{r}_i)$ is the two particle joint probability density. In obtaining the above equation, we have assumed that all the scatterers are identical.

We now assume that the average field $<b^i_{\tau n}>_i$ (the coherent field) propagates in a medium with an effective complex wave number $\vec{K} = (K_1+iK_2)\hat{z}$ in the direction of the original incident field in the discrete random medium:

$$<b^i_{1\sigma m\ell}>_i = i^\ell \, Y_{1\sigma m\ell} \, e^{i \, \vec{K}\cdot\vec{r}_i} \tag{13}$$

$$<b^i_{2\sigma m\ell}>_i = i^\ell \, Y_{2\sigma m\ell} \, e^{i\vec{K}\cdot\vec{r}_i} \; . \tag{14}$$

Substituting Equations (13) and (14) in Equation (12) and invoking the extinction theorem to cancel the incident wave term in (12), we obtain the following equations for the unknown amplitudes $Y_{1\sigma m\ell}$ and $Y_{2\sigma m\ell}$

$$\begin{aligned} i^{n'} Y_{11\ell n'} = \sum_{n=1}^{\infty} \sum_{p=1}^{\infty} \sum_{m=|n-n'|}^{n+n'} & i^p (-i)^m \, I_m \\ & \left\{ Y_{11\ell p} \left[\left(T^{1\ell n}_{1\ell p} \right)^{11} \psi_{11}(n,n',m) + \left(T^{2\ell n}_{1\ell p} \right)^{21} \chi_{21}(n,n',m) \right] \right. \\ & \left. + \, Y_{22\ell p} \left[\left(T^{1\ell n}_{2\ell n} \right)^{12} \psi_{11}(n,n',m) + \left(T^{2\ell n}_{2\ell p} \right)^{22} \chi_{21}(n,n',m) \right] \right\} ; \end{aligned} \tag{15}$$

$\ell \epsilon [0,n']$; $n' \epsilon [1,\infty]$

$$i^{n'} Y_{22\ell n'} = \sum_{n=1}^{\infty} \sum_{p=1}^{\infty} \sum_{m=|n-n'|}^{n+n'} i^{p} (-1)^{m} I_{m}$$

$$\left\{ Y_{11\ell p} \left[\left(T^{1\ell n}_{1\ell p} \right)^{11} \chi_{12}(n,n',m) + \left(T^{2\ell n}_{1\ell n} \right)^{21} \psi_{22}(n,n',m) \right] \right.$$

$$\left. + Y_{22\ell p} \left[\left(T^{1\ell n}_{2\ell p} \right)^{12} \chi_{12}(n,n',m) + \left(T^{2\ell n}_{2\ell p} \right)^{22} \psi_{22}(n,n',m) \right] \right\} \; ; \qquad (16)$$

$\ell \varepsilon [0,n']$; $n' \varepsilon [1,\infty]$

where

$$I_m(K,k,c) = \frac{6c}{(ka)^2 - (Ka)^2} \{2ka \; j_m \; (2Ka) \; h_m'(2ka)$$

$$-2Ka \; h_m(2ka) \; j_m'(2Ka)] + 24c \int_{x=1}^{\infty} x^2 [g(x)-1] \; h_m(kx) \; j_m(Kx) \; dx \qquad (17)$$

$$\psi_{11}(n,n',m) = \psi_{22}(n,n',m) = -i^{n'-n+m} \left[\frac{(2m+1) \;\; (2n'+1}{2n'(n'+1)} \right]$$

$$\left[\frac{n(n+1)}{n'(n'+1)} \right]^{1/2} \left[n(n+1) + n'(n'+1) - m(m+1) \right] \qquad (18)$$

$$\begin{pmatrix} n & n' & m \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} n & n' & m \\ 1 & -1 & 0 \end{pmatrix}$$

$$\chi_{21}(n,n',m) = -\chi_{12}(n,n',m) = -i^{n'-n+m+1}\left[\frac{2(m+1)\ (2n'+1)}{2n'(n'+1)}\right.$$

$$\left.\left[\frac{n(n+1)}{n'(n'+1)}\right]^{1/2}\left[\left\{m^2-(n-n')^2\right\}\left\{(n+n'+1)^2-m^2\right\}\right]^{1/2} \qquad (19)$$

$$\begin{pmatrix} n & n' & m-1 \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} n & n' & m \\ 1 & -1 & 0 \end{pmatrix}.$$

In the above equation, $c = 4\pi n_0 a^3/3$ is the effective spherical concentration. For plane waves propagating parallel to the rotational axis of symmetry of scatterers, only $\ell = 1$ contributes, and also only certain combinations of m yield non-zero T-matrix elements which are used in Equations (15) and (16). In Equation (17), g(x) is the pair correlation function which depends only on $|\vec{x}| = |\vec{\rho}_{ij}|$ due to translational invariance of the system under consideration. To obtain expressions for g(x), a description of the interparticle forces is needed. In our statistics, the dielectric scatterers are assumed to behave like effective hard spheres of radius 'a' where 'a' is the radius of the circumscribing sphere, see Figure 1. Wertheim [1963] has obtained a series solution of the integral equation for the pair correlation function derived by Percus and Yevick [1958] for an ensemble of hard spheres. Throop and Bearman [1965] have used the Wertheim result and provided tabulated values of g(x) as a function of x for several values of c. Plots of g(x) vs x is shown in Figure 2.

At low values of concentration c, $g(x) \approx 1$, see Figure 2 and hence the integral in Equation (17) is negligible which results in a system of uncorrelated hard particles. This is what has been referred to as the well stirred approximation (WSA) and yields the 'hole correction integral' as outlined by Fikioris Waterman [1964] and by us earlier. If $g(x) > 1$, one can regard the

Equation (17) as a modified 'hole correction integral' which is of the same form as used by Twersky [1977, 1978].

Equations (15) and (16) are simultaneous linear homogeneous equations for the unknown amplitudes $Y_{\tau\sigma m i}$. For a nontrivial solution, we require that the determinant of the truncated coefficient matrix C vanishes, which yields an equation for the effective wave number $K = (K_1+iK_2)$ in terms of k and the T-matrix of a scatterer. This is the dispersion relation for the scatterer filled medium. The real part of K relates to the phase velocity while the imaginary part relates to coherent attenuation in the medium.

3. NUMERICAL COMPUTATIONS

In the low concentration limit, $c \to 0$, it is well known that the single scattering approximation (SSA) is valid so that Im(K/k) is given by

$$\mathrm{Im}(K/k) = \frac{3}{8}\, c\, \frac{Q_{ext}}{ka} \tag{20}$$

where Q_{ext} is the normalized (with respect to πa^2) extinction cross section of a sphere of radius 'a'. An important problem is propagation in a rain medium where the single scattering approximation has been widely used. Indeed, even under very heavy rain, the concentration rarely exceeds 0.01 and is typically around 10^{-4}. We have compared our theory using WSA with Equation (20) for a distribution of spherical water drops of radius 0.1 cm with ka in the range $0.1 \leq ka \leq 3$. The refractive index, which is a function of frequency, is taken from Ray [1972]. In Figure (3), we show the attenuation constant γ defined as 4π Im(K)/Re(K) as a function of ka using the WSA for $c = 10^{-2}$, 10^{-3}, and 10^{-4} which is to be compared with Figure (4) which uses SSA. We note that

both solutions yield nearly identical results. In Figure (5), we show computation of γ vs concentration for different ka values using the WSA. Again the SSA is seemed to be excellent for the rain medium.

We now present computations for a random medium model used by Ishimaru [1981] for the optical propagation experiments. The scatterers are latex spheres of diameter 0.107μ immersed in water with incident wavelength $\lambda = 0.6\mu$. In the Rayleigh limit, Twersky [1978b] has given an expression for Im(K/k) by considering the leading effects of the pair-correlation:

$$\mathrm{Im}(K/k) = c(ka)^3 \left[\frac{\varepsilon_r - 1}{\varepsilon_r + 2}\right]^2 W \tag{21}$$

where ε_r is the relative dielectric constant and W is the packing factor given by

$$W = \frac{(1-c)^4}{(1+2c)^2} = 1 + 24c \int_0^\infty x^2[g(x)-1]\, dx \,. \tag{22}$$

In Figure (6), we show Im(K/k) as a function of concentration c using Equation (22) and the present theory employing the WSA, the P-YA and the Matern model. The Matern [1960] model is completely analytic and is valid for $c < 0.125$. We note that Equation (22) and the P-YA are identical while both the Matern model and the WSA fail for $c \approx 0.04$, and in fact they give unphysical results for $c > 0.125$.

In Figure (7), we show the comparison between the computation and the two measured values at $c = 0.01$ and 0.10 given by Ishimaru [1981]. We note that the ka value is 0.56 and that multiple scattering effects are seemed to be important even at $c = 0.01$. The measured values at $c = 0.01$ and 0.1 are in

very good agreement with both the WSA and P-YA while the SSA consistently overestimates the effective coherent attenuation. Also, for $c > 0.10$ where measurements are not available at the present time, we feel that only the P-YA predicts the correct behavior of $\mathrm{Im}(K/k)$. In Figure (8), we show the variation of $\mathrm{Im}(K/k)$ with ka for $c = 0.21$ and compare the results using the SSA, the WSA and the P-YA. Values for the WSA for $ka \leq 0.75$ are not shown since the solution fails $[\mathrm{Im}(K/k) < 0]$ in this region. However, as ka increases it appears that the WSA tends to merge with P-YA for $ka \geq 3.0$. The SSA on the other hand predicts a higher attenuation than either the WSA or the P-Ya.

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ACKNOWLEDGMENTS

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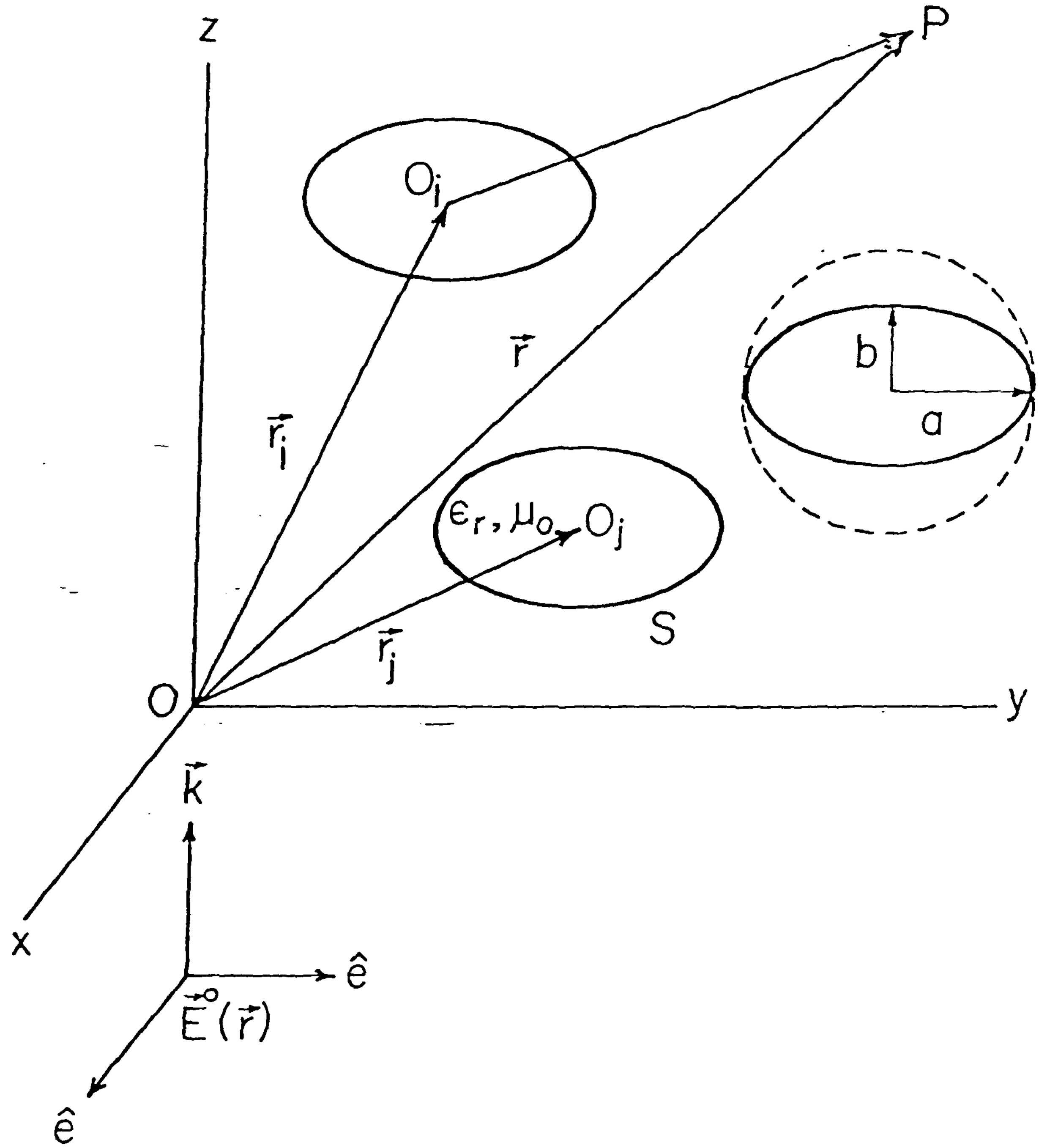

Figure 1. Geometry of randomly distributed and aligned scatterers

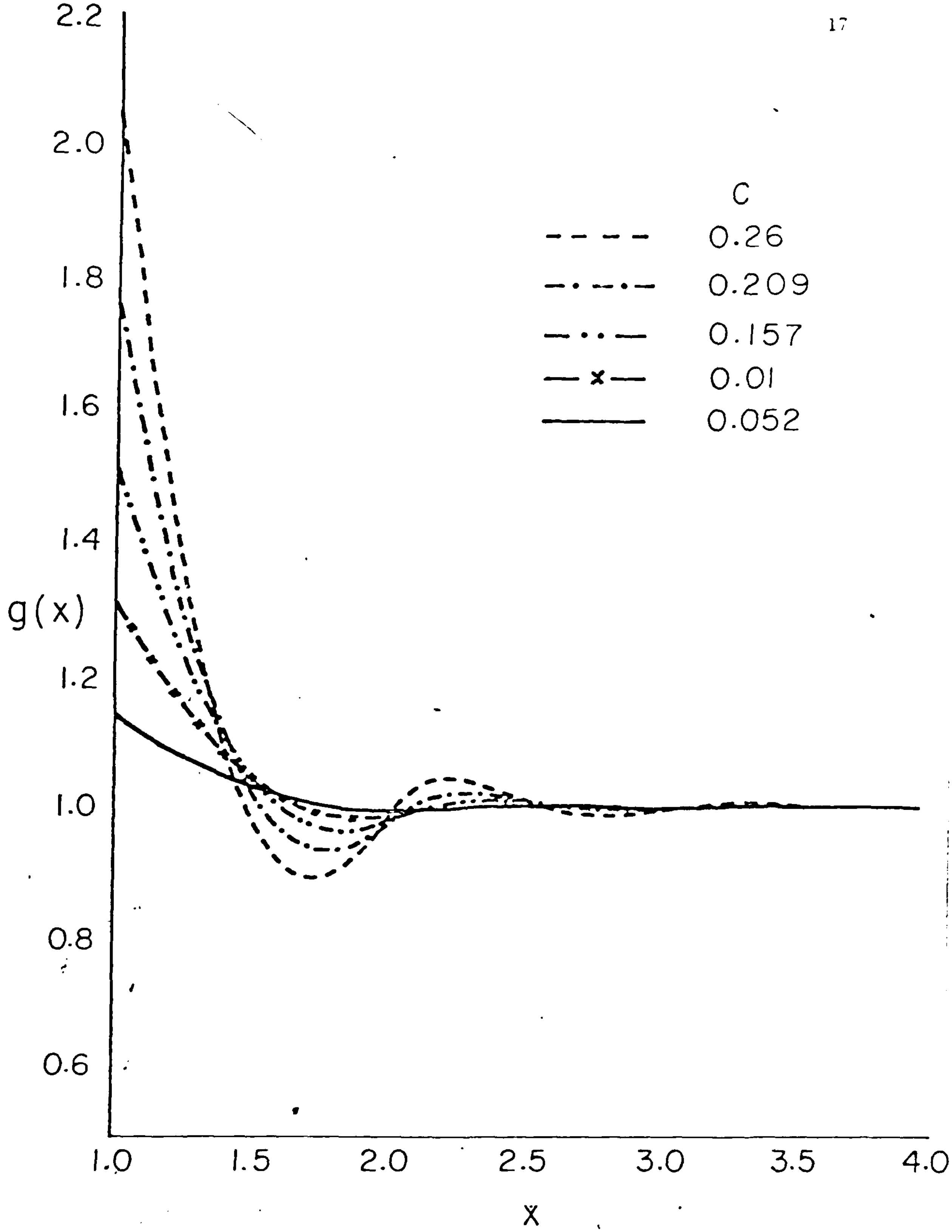

Figure 2. The Percus-Yevick pair correlation function for hard spheres

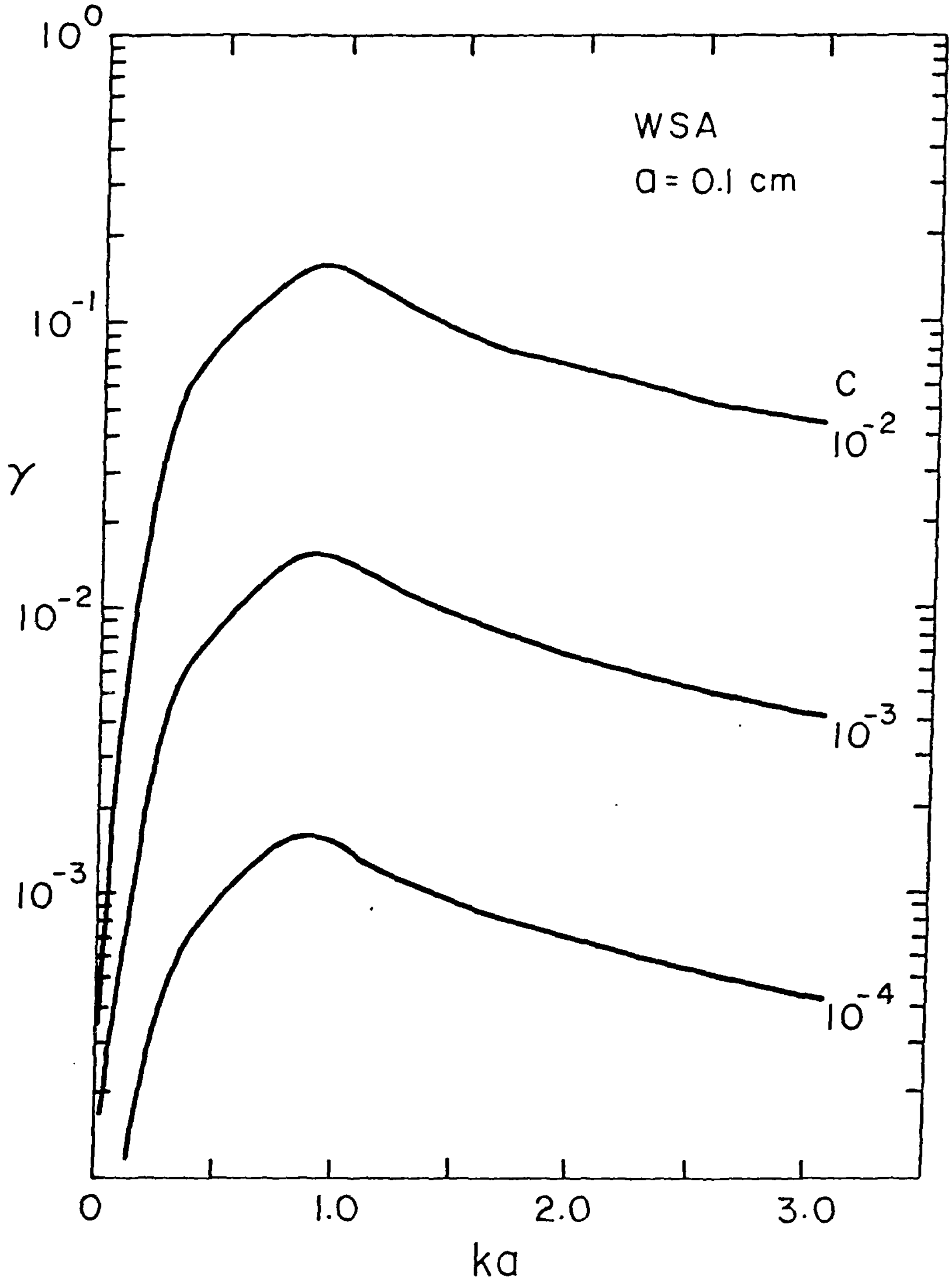

Figure 3. The coherent attenuation constant γ vs ka for $\varepsilon_r = \varepsilon_r(\lambda)$ using the WSA

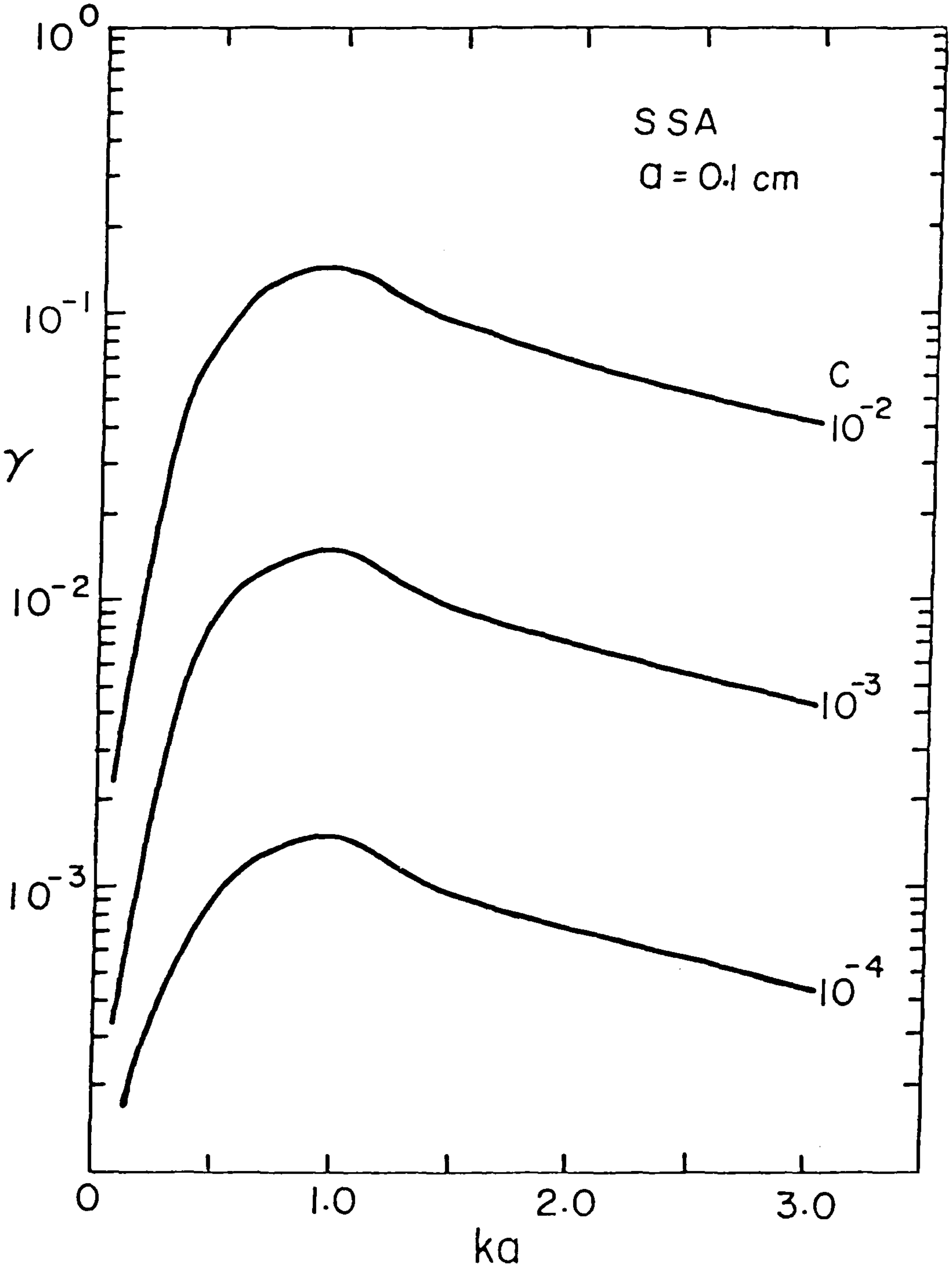

Figure 4. The coherent attenuation constant γ vs ka for $\varepsilon_r = \varepsilon_r(\lambda)$ using SSA

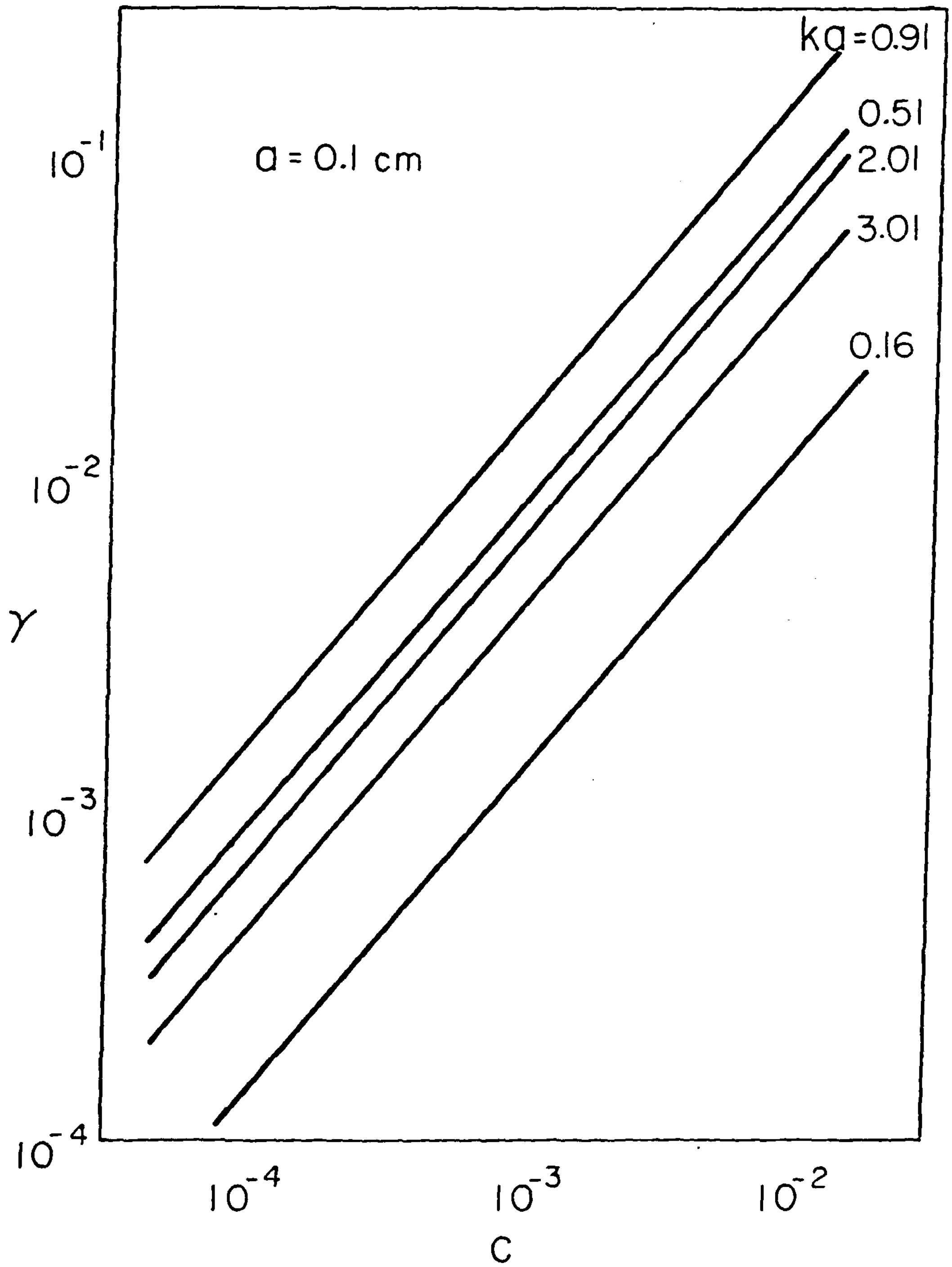

Figure 5. The normalized attenuation constant γ vs concentration c for different values of ka using the WSA

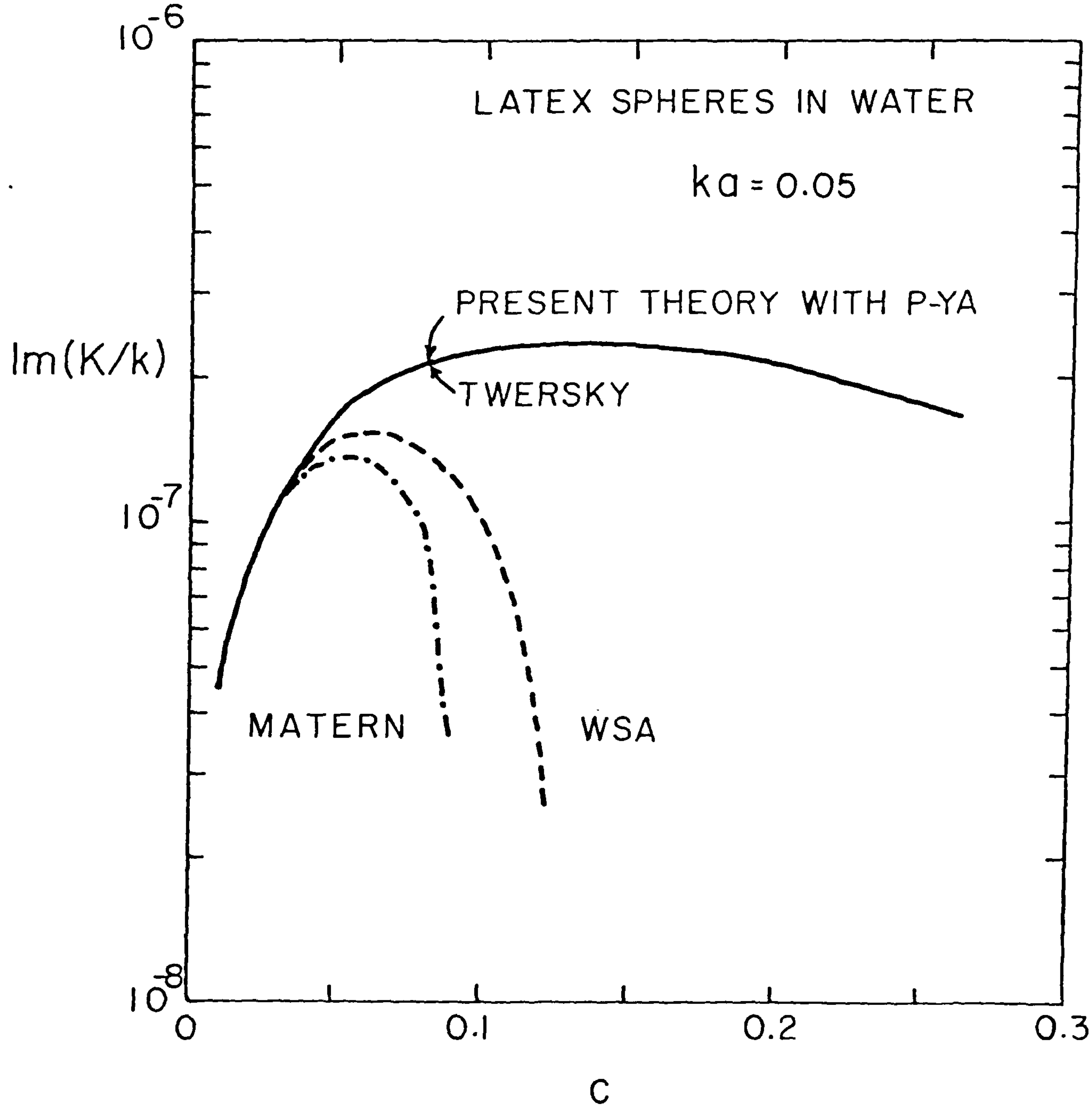

Figure 6. The coherent attenuation Im(K/k) vs concentration c at ka = 0.05 for latex spheres in water

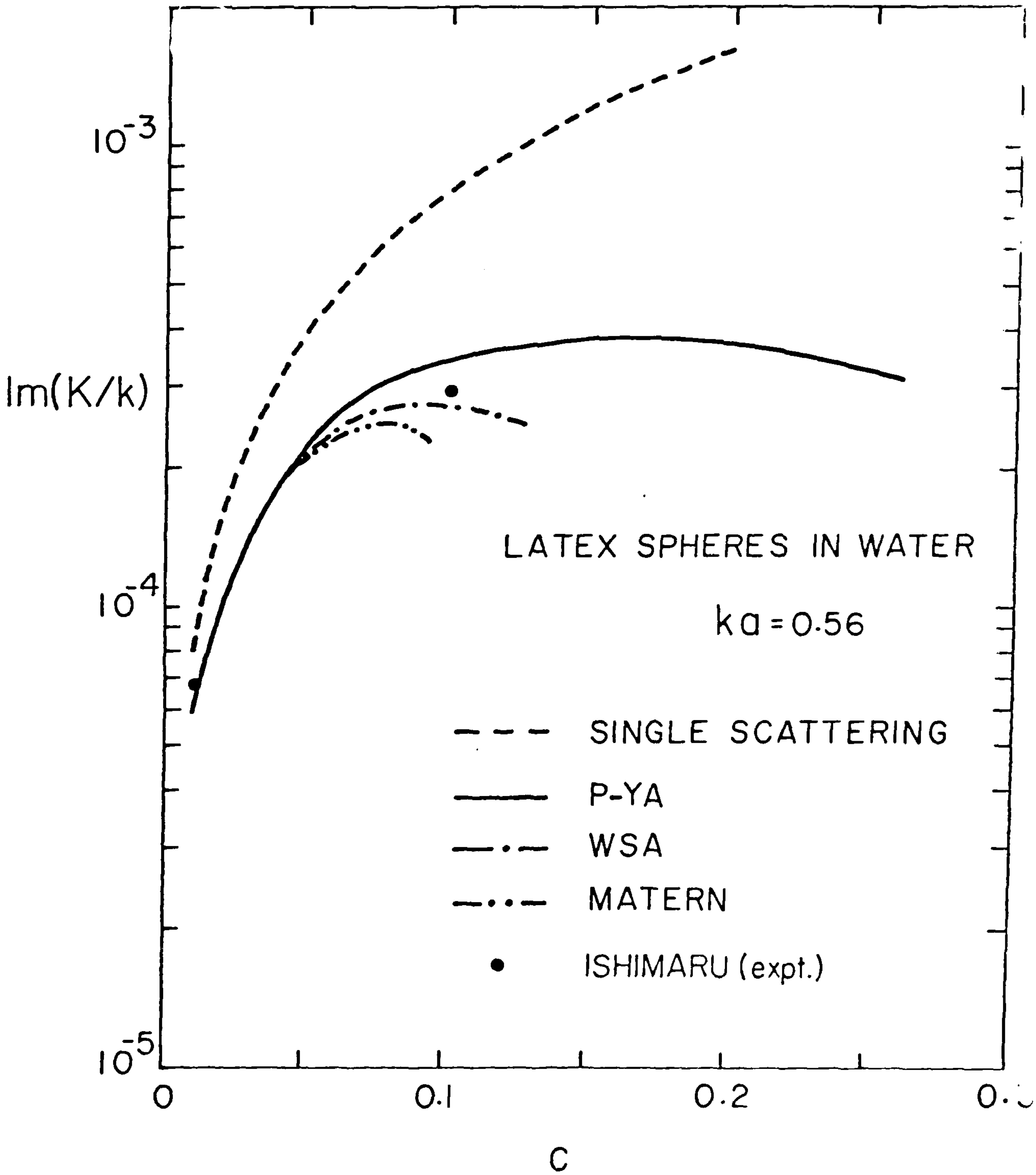

Figure 7. The coherent attenuation Im(K/k) vs concentration c at ka = 0.56 for latex spheres in water using different models of pair correlation functions

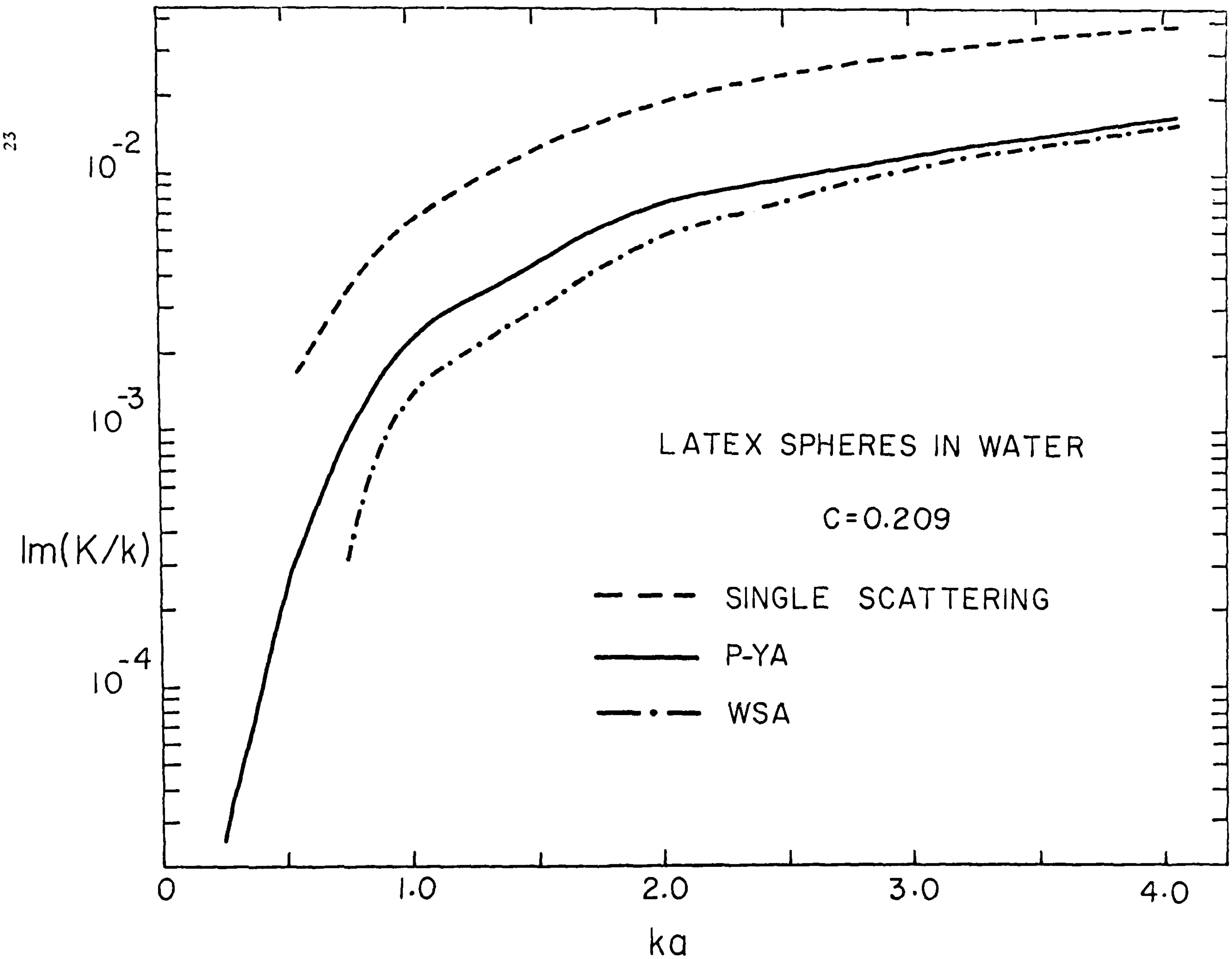

Figure 8. The coherent attenuation Im(K/k) vs ka for c = 0.209 for latex spheres in water using SSA, P-YA and WSA

COHERENT WAVE ATTENUATION AND FREQUENCY DEPENDENT
PROPERTIES OF ABSORBING MATERIALS

by

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Abstract

A scattering matrix theory is presented for studying the multiple scattering of both longitudinal and transverse elastic waves in a medium containing a random distribution of inclusions or voids of arbitrary shape. A statistical analysis with QCA and Percus-Yevick pair correlation function is then employed to obtain expressions for the average amplitudes of the coherent fields which may be solved to yield the bulk or effective properties of the inhomogeneous medium. Suggestions for incorporating CPA in conjunction with QCA so that materials with dense concentration of inclusions can be considered are also given.

Introduction

In recent years, considerable effort has been devoted to promoting the development of elastomeric absorbing materials, containing a distribution of cavities and inclusions, which are bonded to submerged structures to control the sound radiated by these structures as well as to modify their acoustic reflection characteristics (echo reduction). To use such absorbing layers, it is important to determine how their physical properties such as density, thickness and effective elastic moduli, and material composition such as distribution and orientation of the inclusions and their size distributions affect the acoustical behavior of any actual sturcture coated with that material.

The waves incident on such inhomogeneous media undergo multiple scattering due to the presence of inclusions thus reducing the scattering amplitude or cross section by absorption and attenuation of waves. The attenuation depends critically on the material properties of the host medium (matrix) and inclusions, the distribution of the inclusions and the frequency of the incident wave. The problem is very difficult and to our knowledge, rigorous theories with numerical results are not available in the literature.

In multiple scattering theories, approximations are usually made at a very early stage for a) the geometry of the inclusion, b) the size of the inclusion relative to the wavelength of incident wave, and c) distribution of the inclusions in the matrix medium. The approximations with respect to geometry and size are related. If the inclusion is small compared to the incident wavelength, it is not possible to "see" exact

details of the inclusion and usually one is content to obtain the gross scattering properties of the inhomogeneous medium. This is the so-called Rayleigh or low frequency limit, and yields corrections to the solution for point scatterers. As far as the distribution of the inclusions is concerned, one either has regular arrays of inclusions or a random distribution. In the former case, one performs a lattice sum while in the latter case, one employs a configurational averaging procedure. If the concentration of inclusions is small, i.e., the inclusions are sparsely distributed, we may use a single scattering or first Born approximation. Approximations have been employed by many authors and the corresponding effective properties of the medium were studied at the low frequencies and low concentrations, see for example, Waterman and Truell [1], Merkulova [2], Chaban [3,4], Chatterjee and Mal [5], Domany, Gubernatis and Krumhansl [6], Korringa [7], Kröner [8], Datta [9] and the references therein. Actually the real problem warrants a rigorous multiple scattering theory and a computational approach to study the frequency dependent properties of the inhomogeneous media which will be valid for frequencies comparable to scatterer size and for a wide range of concentrations, shapes and sizes.

Recently, the present investigators have developed a multiple scattering formalism by introducing the concept of a T-matrix for individual inclusions that makes the formulation more general and applicable to a variety of different scatterers, see Refs. [10-20]. The method also lends inself to numerical computations for higher frequencies of the incident plane wave as well as more realistic geometries for the inhomogeneities. The dynamic elastic properties of composite elastic media have been studied in [20] using this formulation, and the concept of an average frequency dependent

elastic stiffness tensor following the work of Bedeaux and Mazur [21], and Varadan and Vezetti [22]. The results seem to be promising for future research in this area. In Ref. [20], we have shown that a Clausius-Mosotti type formula for the average shear modulus can be recovered in the low frequency limit. For higher frequencies, we have obtained the dynamic properties for a range of frequencies. The extension of the theory presented in [20] to acoustic and elastic wave scattering will be useful for Naval applications.

The present state of the art is as follows: the statistical considerations seem to be the most difficult for three dimensional inclusions and the least amount of progress has been made in this area. All formalisms that involve ensemble averaging result in a heirarchy of equations for the average fields that involve higher and higher order correlation functions. This heirarchy must be truncated in some fashion. Foldy [23] approximated the field incident on a scatterer by the average field itself. Lax [24] was the first to use a quasi-crystalline approximation which involves the two particle correlation function. At the moment, only the 'hole correction' has been taken into account in a systematic way. Bose and Mal [25] have tried correlation functions that fall off exponentially with distance. Recently, Twersky [26] has used the scaled particle equation of state of a gas of hard spheres to obtain improvements to the hole correction integral. The T-matrix formalism employs Lax's quasicrystalline approximation (QCA), the hole correction integral and results in a set of equations that must be solved in a self-consistant manner.

In this paper, a radially symmetric pair-correlation function given by Percus-Yevick (P-YA) integral equation [27] is introduced which gives

improvements to the hole correction integral. The "well-stirred" approximation (WSA) was used previously by us which assumes no correlation between the scatterers except that they should not interpenetrate. The WSA seems to depend on concentration and frequency. At low frequency or Rayleigh limit, WSA gives good results up to concentration, $c \leq 0.04$ and unphysical results for $c \geq 0.125$ [28], However, at higher frequencies and higher concentration, the WSA with quasi-crystalline approximation (QCA) yields better results. At resonance frequencies we note that P-YA is so far the appropriate correlation function to be employed [29-31].

Formulation of the Problem

Consider N identical, finite elastic inclusions that are randomly distributed in a different elastic medium, see Fig. 1. The scatterers are homogeneous with elastic properties given by Lame's constants λ_1 and μ_1 and density ρ_1. The properties of the outside medium (call matrix) are given by λ, μ and ρ. In Fig. 1, 0_i and 0_j refer to the center of the i-th and j-th scatterers, respectively and they are referred to the origin 0 by the spherical polar coordinates (r_i, θ_i, ϕ_i). P is any point in the medium outside the scatterers (the matrix medium).

A time harmonic plane wave of unit amplitude and frequency ω is incident on the medium such that the direction of propagation of the incident waves is along the z-axis, which may be written in terms of displacement field vector $\vec{u}^0$:

$$\vec{u}^0(\vec{r}) = e^{i(k_p z - \omega t)}\hat{z} + e^{i(k_s z - \omega t)}\hat{x} \qquad (1)$$

where k_p and k_s are the compressional and shear wave numbers given by

$$k_p = \omega/c_p \quad ; \quad c_p = \sqrt{(\lambda+2\mu)/\rho} \tag{2}$$

$$k_s = \omega/c_s \quad ; \quad c_s = \sqrt{\mu/\rho} \tag{3}$$

and t is the time. The waves incident to the discrete random media will undergo multiple scattering. Let $\vec{u}_i^s(\vec{r})$ be the field scattered by the i-th scatterer. The incident and scattered fields satisfy the vector Helmholtz equation. The problem at hand reduces to computing the total wave field at any point in the matrix medium and hence the bulk properties, satisfying the appropriate boundary condition on the surface of the scatterers and radiation conditions at infinity.

The total field at any point in the matrix medium can be interpreted as the sum of the incident field and the fields scattered by all the scatterers, which can be written as

$$\vec{u}(\vec{r}) = \vec{u}^0(\vec{r}) + \sum_{i=1}^{N} \vec{u}_i^s(\rho_i) \quad ; \quad \vec{\rho}_i = \vec{r}-\vec{r}_i \quad . \tag{4}$$

However, the field that excites the i-th scatterer is the incident field $\vec{u}^0$ plus the fields scattered from all other scatterers except the i-th. The term exciting field $\vec{u}^e$ is used to distinguish between the field actually incident on a scatterer and the external incident field $\vec{u}^0$ produced by a source at infinity. Thus, at a point $\vec{r}$ in the vicinity of the i-th scatterer, we write

$$\vec{u}_i^e(r) = \vec{u}^0(\vec{r}) + \sum_{j\neq i}^{N} \vec{u}_j^s(\vec{\rho}_j) \quad ; \quad a \leq |\rho_j| < 2a \tag{5}$$

where 'a' is a typical dimension of the scatterer.

The exciting and scattered fields for each scatterer can be expanded in terms of vector spherical functions with respect to an origin at the center of that scatterer:

$$\vec{u}_i^e(\vec{r}) = \sum_{\tau=1}^{2} \sum_{\ell=1}^{\infty} \sum_{n=0}^{\ell} \sum_{\sigma=1}^{2} b^i_{\tau\ell n\sigma} \, \mathrm{Re}\, \vec{\psi}_{\tau\ell n\sigma}(\vec{\rho}_i) = \sum_{\tau n} b^i_{\tau n} \, \mathrm{Re}\, \vec{\psi}^i_{\tau n} \qquad (6)$$

$$\vec{u}_i^s(\vec{r}) = \sum_{\tau n} B^i_{\tau n} \, \vec{\psi}^i_{\tau n} \qquad (7)$$

where $\vec{\psi}_{\tau\ell n\sigma}$ ($\tau = 1,2,3$) are the vector spherical vector basis functions [19]. Field quantities that are regular at the origin are expanded in terms of the regular (Re) basis set (Re $\vec{\psi}_{\tau\ell n\sigma}$) obtained by replacing the Hankel function of the first kind, h_n, in the above equations by the spherical Bessel functions j_n of the first kind. In Eq. (7), we abbreviate these vector basis functions as $\vec{\psi}_{\tau\ell n\sigma} = \vec{\psi}_{\tau n}$. We note that $\vec{\psi}_{1n}$ is for the longitudinal part while $\vec{\psi}_{2n}$ and $\vec{\psi}_{3n}$ for the transverse parts. The choice of the basis set in Eq. (7) satisfies the radiation condition at infinity for the scattered field, while the choice in Eq. (6) satisfies the regular behavior of the exciting field in the region $a < |\vec{\rho}_i| < 2a$. The superscript i on the basis functions refer to expansions with respect to 0_i, and $b^i_{\tau n}$ and $B^i_{\tau n}$ are the unknown exciting and scattered field coefficients. We also expand the incident field in terms of vector spherical functions:

$$\vec{u}^{0} = \frac{e^{i k_p \zeta_i}}{i k_p} \sum_{s=0}^{\infty} \sum_{t=-s}^{s} (2s+1)\, i^{s}\, \mathrm{Re}\, \vec{\psi}^{i}_{1ts}\, \delta_{t,0}$$

$$+ \frac{1}{2i}\, e^{i k_p \zeta_i} \sum_{s=1}^{\infty} \sum_{t=-s}^{s} \frac{2s+1}{s(s+1)}\, i^{s} \Big\{ \mathrm{Re}\, \vec{\psi}^{i}_{2ts} \left[\delta_{t,1} + s(s+1)\, \delta_{t,-1} \right]$$

$$+ \frac{1}{k_s}\, \mathrm{Re}\, \vec{\psi}^{i}_{3ts} \left[\delta_{t,1} - s(s+1)\, \delta_{t,-1} \right] \Big\} \tag{8}$$

where δ_{mn} is the Kronecker δ. For the sake of simplicity, we write the incident wave field in terms of expansion co-efficients $a_{\tau n}$ as follows

$$\vec{u}^{0} = \sum_{\tau n} a_{\tau n}\, \mathrm{Re}\, \vec{\psi}^{i}_{\tau n}\, e^{i k_\tau \cdot \vec{r}_i} \tag{9}$$

where $a_{\tau n}$ are the known incident field coefficients.

The unknown coefficients $b^{i}_{\tau n}$ can be related to $B^{i}_{\tau n}$ by means of any convenient scattering operator, in this case we employ the T-matrix, see Ref. [32].

$$B^{i}_{\tau n} = \sum_{\tau' n'} T^{i}_{\tau n, \tau' n'}\, b^{i}_{\tau' n'} \quad . \tag{10}$$

Substituting Eqs. (6), (7) and (8) in (5), we obtain

$$\sum_{\tau n} b^{i}_{\tau n}\, \mathrm{Re}\, \vec{\psi}^{i}_{\tau n} = e^{i k_\tau \hat{z} \cdot \vec{r}_i} \sum_{\tau n} \mathrm{Re}\, \vec{\psi}^{i}_{\tau n} + \sum_{j \neq i}^{N} \sum_{\tau n} B^{j}_{\tau n}\, \vec{\psi}^{j}_{\tau n} \tag{11}$$

Since the field quantities are expanded with respect to centers of each scatterer, we obtain Eq. (9) with basis functions with respect to i-th and j-th centers. In order to express them with respect to a common origin 0_i, we employ the translation and addition theorems for the vector spherical functions [33] which may be written in a compact form as follows:

$$\vec{\psi}^{j}_{\tau n} = \vec{\psi}_{\tau n}(\vec{r}-\vec{r}_j) = \sum_{\tau' n'} \sigma_{\tau n\, \tau' n'}\,(\vec{r}_i-\vec{r}_j)\ \mathrm{Re}\ \vec{\psi}^{i}_{\tau' n'} \quad . \tag{12}$$

Employing Eq. (12) in (11) and using the orthogonality of the vector spherical basis functions, we obtain the following set of coupled algebraic equations for the exciting field coefficients $b^i_{\tau n}$

$$b^i_{\tau n} = a_{\tau n}\, e^{i\,\vec{k}_\tau \cdot \vec{r}_i} + \sum_{j\neq i}^{N} \sum_{\tau' n'} B^j_{\tau' n'}\, \sigma_{\tau' n', \tau n}\,(\vec{r}_i-\vec{r}_j) \tag{13}$$

With the scattered field coefficients $B^j_{\tau n}$ expressed in terms of exciting field coefficients $b^j_{\tau n}$ and the T-matrix as given by (10), Eq. (13) gives the exciting field formulation of the multiple scattering. If we multiply both sides of Eq. (13) by the T-matrix, then we obtain the scattered field formulation of multiple scattering which may be written as

$$B^i_{\tau n} \equiv B^{\ell(i)}_{\tau n} = \sum_{\tau'' n''} T^i_{\tau n, \tau'' n''} \Big[a_{\tau'' n''} \exp(i\ \vec{k}_\tau \cdot \vec{r})$$

$$+ \sum_{j\neq i}^{N} \sum_{\tau' n'} B^j_{\tau' n'}\, \sigma_{\tau' n', \tau'' n''}\,(\vec{r}_i-\vec{r}_j) \Big] \quad . \tag{14}$$

From Eq. (14), it can be seen that the scattered field coefficients of the i-th scatterer explicitly depend on the position and orientation of other scatterers. In this paper, we consider a random distribution of spherical scatterers and the case when $N \to \infty$ and the volume occupied by the scatterers $V \to \infty$ such that $N/V = n_0$ is a finite number density. For such distribution, a configurational average of Eq. (14) can be made over the positions of all scatterers [28-32] with QCA [24] to arrive at an equation for the configurational average $\langle B^{i}_{\tau n}\rangle_i$ of the scattered field coefficients with one scatterer fixed:

$$\langle B^{i}_{\tau n}\rangle_i = \sum_{\tau'' n''} T_{\tau n,\tau'' n''} \Bigg[a_{\tau'' n''}\, e^{i \vec{k}_\tau \cdot \vec{r}_i} + (N-1) \sum_{\tau' n'} \int_V p(\vec{r}_j|\vec{r}_i) \langle B^{j}_{n}\rangle_j\, \sigma_{\tau' n',\, \tau'' n''}\, d\vec{r}_j \Bigg] \qquad (15)$$

where $p(\vec{r}_j|\vec{r}_i)$ is the two particle joint probability density.

The joint probability density is defined as

$$p(\vec{r}_j|\vec{r}_i) = \begin{cases} \frac{1}{V}\, g(|\vec{r}_j-\vec{r}_i|) & ; \quad |\vec{r}_j-\vec{r}_i| \geq 2a \\ 0 & ; \quad |\vec{r}_j-\vec{r}_i| > 2a \end{cases}$$

Equation (16) implies that the particles are hard (no-interpenetration) and the excluded volume is a sphere of radius 'a' although the particles themselves may be non-spherical. The function $g(|\vec{r}_j-\vec{r}_i|)$ is called the pair correlation function and depends only on $|\vec{r}_j-\vec{r}_i|$ due to

translational invariance of the system under consideration. The pair correlation function for an ensemble of particles depends on the nature and range of the interparticle forces. The average of several measurements of a statistical variable that characterizes an ensemble will depend on the pair correlation function. To obtain expressions for the pair correlation function, one needs a description of the interparticle forces. In our case we assume that the scatterers behave like effective hard spheres (where the radius 'a' is that of the sphere circumscribing the scatterer). Percus and Yevick [27] have obtained an approximate integral equation for the pair correlation function of a classical fluid in equilibrium. Wertheim [34] has obtained a series solution of the integral equation for an ensemble of hard spheres. The statistics of the fluid are then same as those of the ensemble of discrete hard particles that we are considering.

Although integral expressions for the correlation functions also result in a heirarchy, Percus and Yevick have truncated the heirarchy by making certain approximations that result in a self-consistent relation between the pair correlation function $g(x)$ and the direct correlation function $C(x)$. The direct correlation function may be interpreted as the correlation function resulting from an 'external potential' that produces a simultaneous density fluctuation at a point and the external potential is taken to be the potential seen by a particle given that there is a particle fixed at another site. Fisher [35] comments that the Percus-Yevick approximation is a strong statement of the extremely short range nature of the direct correlation function. The integral equation has the form

$$\tau(x) = 1 + n_o \int_{x<2a} \tau(x')dx' - n_o \int_{\substack{x'<2a \\ |x-x'|>2a}} \tau(x')\tau(x-x')dx' \qquad (17)$$

where

$$\begin{aligned} \tau(x) &= g(x) \quad ; \; x > 2a \\ g(x) &= 0 \quad ; \; x < 2a \\ \tau(x) &= -C(x) \quad ; \; x < 2a \\ C(x) &= 0 \quad ; \; x > 2a \end{aligned} \qquad (18)$$

Wertheim [34] has solved the integral equation by Laplace transformation that results in an analytic expression for C(x) in the form

$$C(x) = -(1-\eta)^{-4}\,[(1+2\eta)^2 - 6\eta(1+\tfrac{1}{2}\eta)^2 x + \eta(1+2\eta)^2\, x^3/2] \;;\; \eta = c/8 \qquad (19)$$

where 'c' is the effective spherical concentration of the particles. The Percus-Yevick approximation fails as the concentration approaches the close packing factor for spheres and is expected to be good for $c < 0.3$ or 0.4.

Equation (19) can be substituted back into Eq. (17) to yield a series solution for g(x) in the form [34]

$$g(x) = \sum_{n=1}^{\infty} g_n(x) \qquad (20)$$

where

$$g_n(x) = \frac{1}{24\eta x i} \int e^{t(x-n)}\, [L(t) \mid S(t)]^n\, t\,dt \qquad (21)$$

where

$$S(t) = (1-\eta^2)t^3 + 6\eta(1-\eta)t^2 + 18\eta^2 t - 12\eta(1+2\eta) \tag{22}$$

and

$$L(t) = 12\eta\ [(1+\eta 2)t + (1+2\eta)]\ . \tag{23}$$

Throop and Bearman [36] have tabulated $g(x)$ as a function of x for values of $\eta = c/8$. A few representative plots of the pair correlation function are shown in Fig. 2.

To solve the integral equations given by (15), we consider the inhomogeneous medium with discrete scatterers as a homogeneous continuum and assume that the average coherent wave is a plane wave propagating with an effective wave number K in the same direction as the incident plane wave. We can thus write

$$\langle B^i_{\tau n} \rangle = X_{\tau n}\, e^{i\, \vec{K}\cdot\vec{r}_i} \tag{24}$$

where $X_{\tau n}$ is the amplitude of the coherent wave.

Substituting Eq. (24) in (15) employing the joint probability function as defined before and the divergence theorem to convert the volume integral in (15) to surface integrals and using the extinction theorem which cancels the incident wave, we obtain a set of simultaneous coupled homogeneous equations for the coefficients $X_{\tau n}$ given by

$$X_{\tau n} = c \sum_{\tau'' n''} \sum_{\tau' n'} \sum_{q=|n'-n''|}^{|n'+n''|} X_{\tau' n'}\, T_{\tau n,\tau'' n''}\, C^q_{\tau' n',\tau'' n''}\, \frac{I_q}{(k^2_{\tau'} - K^2)a^2} \tag{25}$$

where $c = 4\pi a^3 n_0/3$ is the effective spherical concentration of the scatterers per unit volume, C^q is an expression containing Wigner coefficients, and

$$I_q(K,k_\tau,c) = \frac{6c}{(k_\tau a)^2-(Ka)^2} \; [2k_\tau a \; j_q(2Ka) \; h_q'(2k_\tau a)$$

$$-2Ka \; h_q(2k_\tau a) \; j_q(2Ka)] + 24c \int_{x=1}^{\infty} x^2[g(x)-1] \; h_q(k_\tau x) \; j_q(Kx) \; dx \qquad (26)$$

At low values of concentration c, $g(x) \gtrsim 1$, see Fig. 2, and hence the integral in Eq. (26) is negligible which results in a system of uncorrelated hard particle statistics. This is what has been referred to as the 'well stirred approximation' (WSA) and yields the 'hole correction integral' as outlined by Fikioris and Waterman [37] and by us earlier. If $g(x) > 1$, one can regard the Eq. (26) as a modified 'hole correction integral' which is of the same form used by Twersky [26].

Equation (25) is a system of simultaneous linear homogeneous equations for the unknown amplitudes $X_{\tau n}$. For nontrivial solution, we require that the determinant of the truncated coefficient matrix vanishes, which yields an equation for the effective wave number K in terms of k_τ and the T-matrix of the scatterer. This is the dispersion relation for the scatterer filled medium. Equation (25) is a general expression valid for any arbitrary shaped scatterer, since the T-matrix is the only factor that contains information about the exact shape and boundary conditions at the scatterer. Thus the formalism presented here is valid for all the three wave fields. The effective wave number K obtained in the analysis is a

complex quantity, the real part of which relates to the phase velocity, while the imaginary part relates to attenuation of coherent waves in the medium.

Results and Conclusions

In the Rayleigh or low frequency limit, the size of the scatterers is considered to be small when compared to the incident wavelength. It is then sufficient to take only the lowest order coefficient in the expansion of the fields. In this limit, the elements of the T-matrix can be obtained in closed form for various simple shapes (46). It can be shown that at low frequencies, only $X_{\tau 0}$, $X_{\tau 1}$ and $X_{\tau,-1}$ of Eq. (25) make a contribution. After some manipulations of the resulting 3 x 3 determinant, we obtain the following dispersion relations for elastic spherical inclusions embedded in a different elastic medium (matrix):

$$\left(\frac{K_p}{k_s}\right)^2 = \frac{(1+9c\, E_1)(1+3c\, E_0)\left[1+3c\, \frac{E_2}{2}\left(2+\frac{3k_s^2}{k_p^2}\right)\right]}{1-15c\, E_2\left[1+3c\, E_0\right]+\frac{3}{2}\, c\, E_2\left(2+\frac{3k_s^2}{k_p^2}\right)} \tag{27}$$

$$\left(\frac{K_s}{k_s}\right)^2 = \frac{(1+9c\, E_1)\left(1+\frac{3}{2}\, c\, E_2\left[2+\frac{3k_s^2}{k_p^2}\right]\right)}{1+\frac{3}{4}\, c\, E_2\left(4-\frac{9k_s^2}{k_p^2}\right)} \tag{28}$$

where

$$E_0 = \frac{1}{3}\ \frac{3\lambda+2\mu-(3\lambda_1+2\mu_1)}{4\mu+3\lambda_1+2\mu_1}$$

$$E_1 = \frac{1}{9}\ (\frac{\rho_1}{\rho} - 1) \tag{29}$$

$$E_2 = -\ \frac{\frac{4}{3}\ \mu(\mu_1-\mu)\ \ 24\mu_1(\mu_1-\mu)\ -\ (\lambda_1+2\mu_1)\ \ (19\mu_1+16\mu)}{24\mu_1(\mu_1-\mu)\ -\ (\lambda_1+2\mu_1)\ \ (19\mu_1+16\mu)\ .}$$

$$x\ \frac{1}{4\mu(\mu_1-\mu)\ +\ 3(\lambda+2\mu)\ \ (2\mu_1+3\mu)}$$

and $c = 4\pi a^3 n_0/3$ is the concentration of spheres, and K_p and K_s are the coherent wave numbers for longitudinal and shear waves, respectively, in the new medium. Similar expressions can also be derived for spheroidal inclusions using the T-matrix obtained in Refs. [32,38]. In the Rayleigh limit, the value of K as determined by the above dispersion relations is a real quantity for lossless (elastic) material and a complex quantity for lossy (viscoelastic) material, and relates to phase velocity $V_p = \omega/K$. In this limit, we normally study the dependence of phase velocity on concentration, angle of incidence and aspect ratio of [illegible] scatterers. The teneral tendency of the phase velocity is to in[illegible] [illegible]ghtly (for inclusion) and decrease slightly (for cracks and cavities) as concentration increases. Thus, the phase velocity vs. concentration information is not very useful both from theoretical and experimental point of view. The

plots of absorption and coherent attenuation due to multiple scattering vs. frequency for various concentrations carry more information which may eventually be used for designing absorbing materials [39].

The dispersion relations given in Eqs. (27) and (28) may also be useful in obtaining the effective shear modulus and bulk modulus at low frequencies. Following the work by us [20,22] and by Bedeaux and Mazur [21], we arrive at the following shear and bulk moduli ($\langle\mu\rangle$ and $\langle B\rangle$) of an elastic material containing a random distribution of stress free bubbles or cavities

$$\frac{\langle\mu\rangle}{\mu} = \frac{4\mu-3c\ E_2\ (9\lambda+14\mu)}{4\mu+6c\ E_2\ (3\lambda+8\mu)} \tag{30}$$

$$\frac{\langle B\rangle}{B} = \frac{3\lambda+2\mu\ [1-6c\ E_0]}{(3\lambda+2\mu)\ [1+3c\ E_0]} \tag{31}$$

where E_0 and E_2 are defined in Eq. (29).

To study the response at resonant and higher frequencies, we must consider higher powers of $k_\tau a$, and this implies that a larger number of terms ($X_{\tau n}$) must be kept in the expansion of the average field. This is best done numerically. For a given value of ka, the T-matrix for the scatterer is computed. Next, the coefficient matrix M corresponding to $X_{\tau n}$ (Eq. (25) is formed. The complex determinant of the coefficient matrix is computed using standard Gauss elimination techniques. For a given $k_\tau a$, the root of the equation det M = 0 is searched in the complex K plane ($K_1 + iK_2$) using Muller's method. Good initial guesses were provided by the Rayleigh limit solutions at low values of $k_\tau a$ and

these could be used systematically to obtain convergence of roots at increasingly higher values of $k_\tau a$. The real part K_1 determines the phase velocity, while the imaginary part K_2 determines the coherent wave attenuation.

Here, we present some sample numerical calculation of spherical glass inclusion in epoxy matrix. The longitudinal and shear wave velocities of the glass and epoxy matrix are taken as $(c_p)_1 = 5.28$ mm/μsec, $c_p = 2.54$ mm/μsec, $(c_s)_1 = 3.24$ mm/μsec and $c_s = 1.16$ mm/μsec, respectively. We consider a concentration of 44.1% to reflect a high concentration. The coherent wave attenuation vs. frequency (longitudinal wave number) for this configuration is shown in Fig. 3. The general tendency of attenuation is to increase at lower frequencies and shows some oscillation as shown. These results are compared with some experimental observations for the same composite obtained by Kinra (private communication). The theoretical results obtained in this paper compare with Kinra's experimental results qualitatively not quantitatively. The reason for this factor difference must be explored in the future. The oscillation at higher frequencies, however, indicate that the scattering is mostly in the forward direction. Thus, in this case repeated scattering should not be important, since the backscattered wave is significantly smaller than the forward scattered wave. The same observation may be noticed even for electromagnetic waves [28] where the theoretical results obtained by our theory are compared with experimental data. (The paper [28] is enclosed for the benefit of the reader.

Since the phase velocity does vary very slightly as a function of frequency, the bulk properties depend totally on coherent wave attenuation

only. Thus, one can compute the bulk properties which can be plotted in the complex plane (Cole-Cole plot) as shown in our paper [30] which is also enclosed.

Recommendations for Future Work

It is obvious from the preceding discussions on the QCA as well as the numerical results that the two major improvements required are for the QCA as well as the pair correlation function, so that good results can be obtained for all concentrations even at long and intermediate values of the wavelength. In a review article, Lax [40] has suggested that in the quantum mechanical context, the QCA could be improved by using modified propagators for the fields. In the classical context, this implies that on the average, single particle scattering takes place in a macroscopically homogeneous medium, and, in this respect, this idea is the same as the coherent potential approximation (CPA) of Solid State Physics. The repeated multiple scattering between pairs of scatterers or cluster effects can be improved by making the self consistent approximation (SCA) in addition to the CPA.

For the purpose of discussion of these ideas within the T-matrix formalism given earlier, we denote by $\vec{u}_j^s$ and $\vec{u}_j^e$ the fields scattered by and exciting the j-th scatterer, respectively. The expansion coefficients of these fields are denoted by B^j and b^j, respectively, omitting all subscripts.

The CPA can be expressed succinctly as

$$\langle B^j \rangle_j = T(K) \langle b^j \rangle_j \tag{32}$$

where the T-matrix relating the exciting and scattered field coefficients is evaluated using the bulk propagation constant K for the embedding medium. Thus the CPA implies that the field scattered by a single obstacle in the presence of several others when averaged over the position of all scatterers is the same as the field that would be produced by a single particle embedded in a macroscopically homogeneous medium described by the propagation constant K. The incorporation of the CPA into the previous formalism involves changes only in the computations and a redefinition of the T-matrix in Eq. (10). It would be interesting to see the change, if any, in the numerical computations as a result of invoking the CPA.

The idea behind the 'self consistent approximation' (SCA) is somewhat more suble. From the discussion in the section on the QCA, it is now clear that QCA-CPA neglects *multiple scattering* between two fixed scatterers. The SCA as defined by Schwartz and Ehrenreich [41] restores this by stating that

$$\langle B^j \rangle_{ij} = T(K) \langle b^j \rangle_j \tag{33}$$

where T(K) is the T-matrix of scatterer 'j' in the presence of scatterer 'i' in the effective medium with propagation constant K. Expressions for T(K) as given by Varadan and Varadan [42] may be written as

$$T(K) = R(\vec{r}_{ij}/2) \quad T[1-\sigma(-\vec{r}_{ij})T\sigma(r_{ij})T]^{-1}$$
$$[1+\sigma(-\vec{r}_{ij})TR(\vec{r}_{ij})] \quad R(-\vec{r}_{ij}/2) \tag{34}$$

where σ is a compact notation for the translation matrices B and C introduced in Eq. (12). The R matrix is simply the part of σ that is regular at the origin, i.e., for $|\vec{r}_{ij}| = 0$. All matrices in Eq. (34) are obtained using the bulk Propagation Constant for the host medium.

We observe that T(K) explicitly depends on r_{ij} the distance between 'i' and 'j'. The integration procedure will no longer be simple as before and the SCA may be rather difficult to enforce in computations, especially if more realistic models are chosen for the pair correlation function.

Incc·poration of the CPA as well as improved models of the pair correlation function into our computations are in progress. We hope that they will shed some light on the sensitivity of multiple scattering theories to approximations like QCA and SCA as a function of frequency and scatterer concentration. Needless to say additional experimental results are required for comparison with these computations.

Acknowledgments

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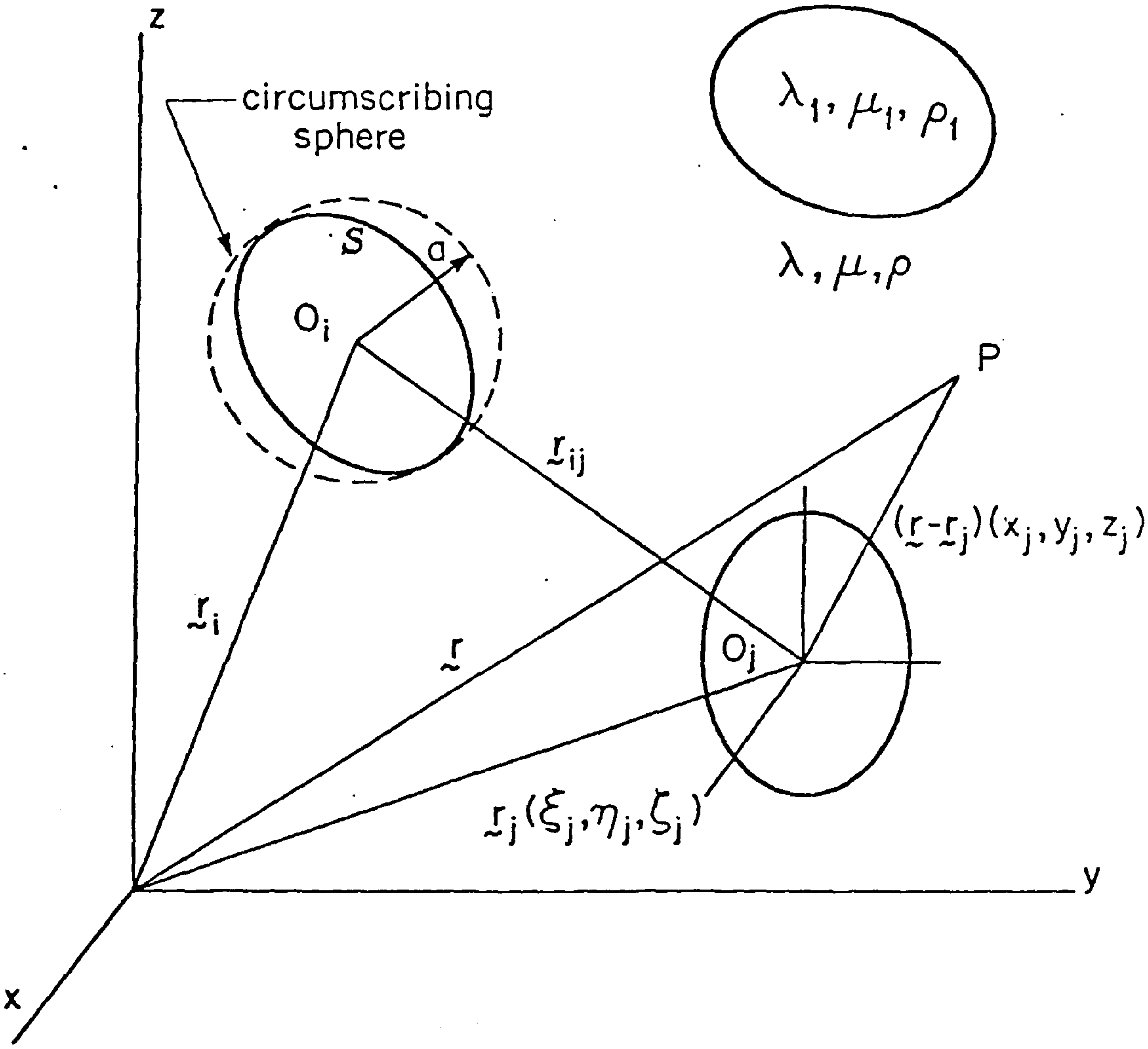

Figure 1. Random distribution of inclusions of arbitrary shape

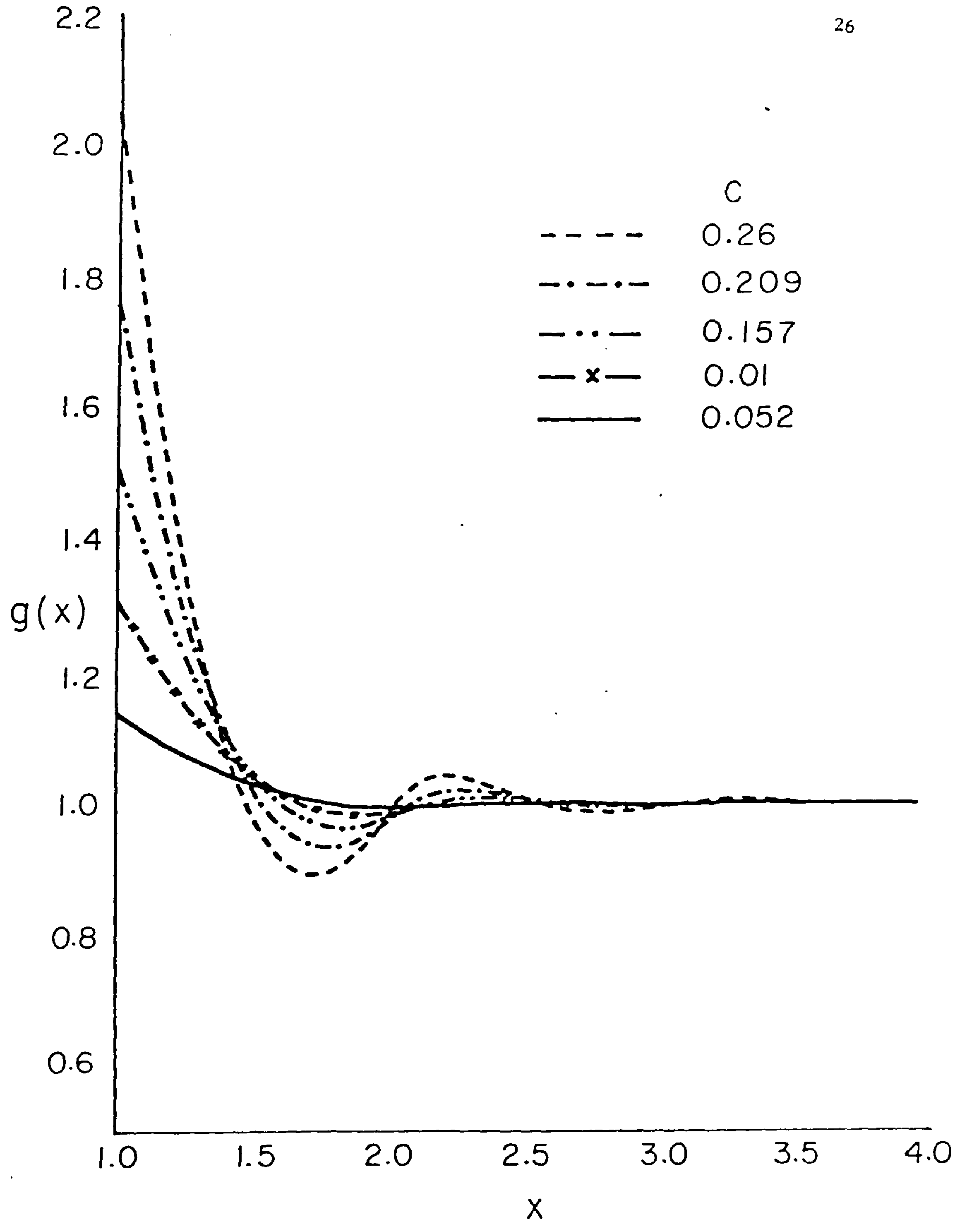

Figure 2. The Percus-Yevick pair correlation function for hard spheres as given by Throop and Bearman (1964)

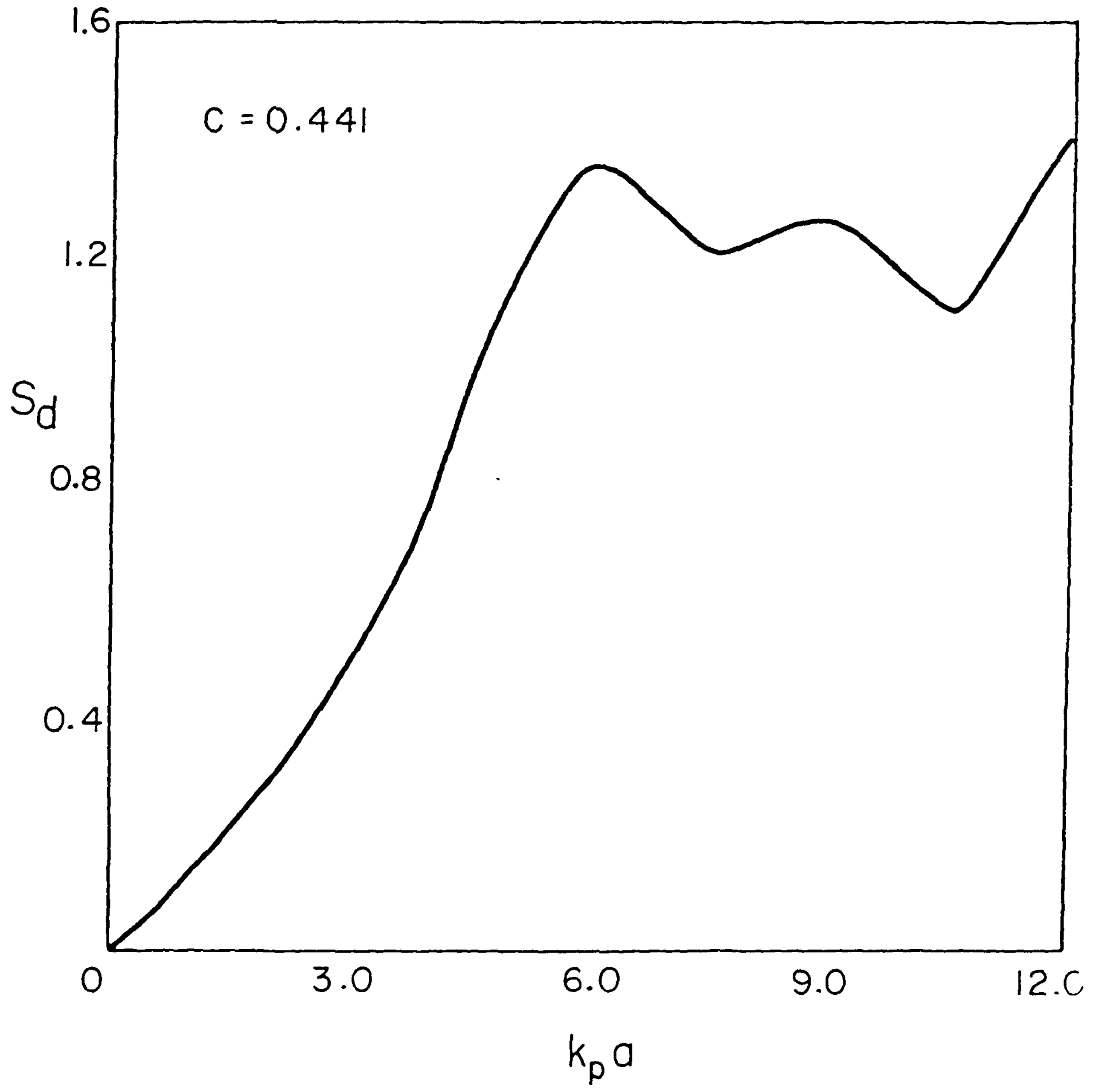

Figure 3. The coherent attenuation $S_d = 4\pi\ \mathrm{Im}(K_\tau/k_\tau)$ vs $k_p a$ for glass spheres in epoxy matrix

FREQUENCY DEPENDENT DIELECTRIC CONSTANTS

OF DISCRETE RANDOM MEDIA

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Abstract

Numerical computations of the effective dielectric constant of discrete random media are presented as a function of frequency. Such media have a complex dielectric constant giving rise to absorption of a propagating wave both due to geometric dispersion or multiple scattering as well as absorption, if any, due to the viscosity of the particles and the matrix medium. We are concerned with the absorption due to multiple scattering. The scattering characteristics of the individual particles are described by a transition or T-matrix. The effects of two models of the pair correlation function which arises in the multiple scattering analysis are considered. We conclude that the well stirred approximation (WSA) is good for sparse concentrations and/or high frequencies whereas the Percus-Yevick approximation (P-YA) is preferred for higher concentrations.

Introduction

The study of the frequency dependence of the effective dielectric constant of statistically inhomogeneous media is important for practical applications such as geophysical exploration, artificial dielectrics etc. In such dielectrics a propagating electromagnetic wave undergoes dispersion and absorption. Some materials are naturally absorptive due to viscosity whereas inhomogeneous media exhibit absorption due to geometric dispersion or multiple scattering.

In this paper the effective, complex frequency dependent dielectric constant of a discrete random medium containing a distribution of aligned spheroidal dielectric scatterers in free space is calculated for different concentrations of the scatterers as well as for different material properties of the scatterers. We use a multiple scattering formalism analogous to that used by Twersky[1] but use the concept of a transition matrix or T-matrix to characterize the scattering from a single obstacle. All details of the geometry and material properties of the scatterer are contained in the T-matrix leaving the general formalism independent of the type of scatterer. Spherical statistics are used even though the scatterers may be non-spherical. Lax's[2] quasi-crystalline approximation (QCA) is used to truncate the heirarchy of equations that result when an ensemble average is performed on the multiply scattered field.

The resulting equations for the average field require a knowledge of the pair correlation function of the dielectric scatterers. In previous work[3,4,5], we assumed that the particles did not penetrate each other but were otherwise uncorrelated. Willis[5] has called this the well stirred approximation (WSA). However, the WSA lead to unphysical results for the absorption coefficient of the average medium for scatterer concentrations $c > 0.125$. In many artificial dielectrics, the scatterer concentration is often greater than 0.125. In this paper, we have also considered the Percus-Yevick[7] approximation (P-YA) to the pair correlation function. Wertheim[8] has provided a semi-analytical solution of the resulting integral equation for a system of hard spheres. Throop and Bearman[9] have provided tabulated values of the pair correlation function for different values of the concentration as a function of the inter particle distance. We have used these tabulated values in the numerical computations.

Calculations are presented for a system of polyethylene spheres and spheroids as well as ice particles for $0 < c < 0.26$ for several values of the non-dimensional wavenumber $ka = \frac{\omega a}{c}$ ranging from 0 to 5.0. ('a' is a characteristic dimension of the scatterer). Two types of results are presented. In the first instance the validity of the WSA and P-YA and their effect on the absorption coefficient is studied as a function of concentration and frequency. Secondly, the complex plane locus of the effective dielectric constant is plotted for the systems considered. For artificial dielectrics the locus deviates dramatically from the circular arc locus commonly noticed for ordinary solids and liquids that exhibit absorption due to viscosity.

Wave propagation in a discrete random medium

Consider N identical rotationally symmetric dielectric scatterers that are aligned but distributed randomly in free space (see Fig. 1). Let O be the origin of a coordinate system located outside the scatterers whose centers are denoted by $O_1, O_2, O_3 \ldots O_N$. Monochromatic plane electromagnetic waves of frequency ω propagate along the symmetry axis of the scatterers which is taken to be the z-axis. Since the medium is isotropic about the z-axis there are no depolarization effects. The time dependence of the incident and hence the fields scattered by the individual scatterers is all of the form $\exp(-i\omega t)$ and this is suppressed in the equations that follow.

Let $\vec{E}_o(\vec{r})$ be the electric field arising from the incident plane wave and $\vec{E}^s_i(\vec{r})$ the field scattered by the i-th scatterer. The total field at a point $\vec{r}$ outside all the N scatterers, denoted by $\vec{E}(\vec{r})$ is given by

$$\vec{E}(\vec{r}) = \vec{E}^o(\vec{r}) + \sum_{i=1}^{N} \vec{E}^s_i(\vec{r})$$

The field incident on or exciting the i-th scatterer is given by

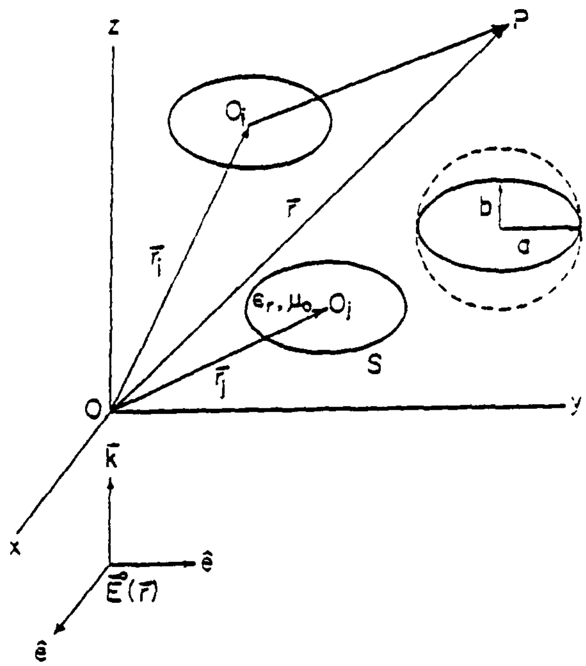

Figure 1. Scattering geometry

$$\vec{E}^e_i(\vec{r}) = \vec{E}^o(\vec{r}) + \sum_{j \neq i}^{N} \vec{E}^s_j(\vec{r}) \; ; \; a \leq |\vec{r}-\vec{r}_i| < 2a \tag{2}$$

where 'a' is a typical dimension of the scatterer. From Eqs. (1) and (2) we note that

$$\vec{E}(\vec{r}) = \vec{E}^e_i(\vec{r}) + \vec{E}^s_i(\vec{r}) \tag{3}$$

We need an additional equation relating $\vec{E}^e_i$ and $\vec{E}^s_i$ in order to make the fields microscopically self-consistent.

Vector spherical functions are used to expand the exciting and scattered fields associated with each scatterer with respect to an origin at the center of that scatterer. Thus

$$\vec{E}^e_i(\vec{r}) = \sum_{\tau=1}^{2} \sum_{l=1}^{\infty} \sum_{m=0}^{l} \sum_{\sigma=1}^{2} A^i_{\tau l m \sigma} \mathrm{Re}\, \vec{\psi}_{\tau l m \sigma}(\vec{\rho}_i) \; ; \; a \leq |\vec{\rho}_i| < 2a \tag{4}$$

and

$$\vec{E}^s_i(\vec{r}) = \sum_{\tau=1}^{2} \sum_{l=1}^{\infty} \sum_{m=0}^{l} \sum_{\sigma=1}^{2} F^i_{\tau l m \sigma} \mathrm{Ou}\, \vec{\psi}_{\tau l m \sigma}(\vec{\rho}_i) \; ; \; |\vec{\rho}_i| \geq 2a \tag{5}$$

where

$$\vec{\rho}_i = \vec{r}-\vec{r}_i \tag{6}$$

and the vector spherical functions are defined as

$$\left\{\begin{matrix}\text{Ou}\\ \text{Re}\end{matrix}\right\} \vec{\psi}_{1\ell m\sigma}(\vec{r}) = \nabla\times \left\{\vec{r}\begin{matrix} h_\ell(kr)\\ j_\ell(kr)\end{matrix}\right\} Y_{\ell m\sigma}(\theta\phi) ; \tag{7}$$

$$\left\{\begin{matrix}\text{Ou}\\ \text{Re}\end{matrix}\right\} \vec{\psi}_{2\ell m\sigma}(\vec{r}) = \frac{1}{k}\nabla\times \left\{\begin{matrix}\text{Ou}\\ \text{Re}\end{matrix}\ \vec{\psi}_{1\ell m\sigma}(\vec{r})\right\} \tag{8}$$

In Eqs. (7) and (8) j_ℓ and h_ℓ are the spherical Bessel and Hankel functions and the $Y_{\ell m\sigma}(\theta,\phi)$ are the normalized spherical harmonics defined with real angular functions. To make the notation more compact we introduce a super index 'n' to represent $(\tau\ell m\sigma)$ as follows

$$\begin{matrix}\text{Ou}\\ \text{Re}\end{matrix}\ \vec{\psi}_{\tau\ell m\sigma} = \begin{matrix}\text{Ou}\\ \text{Re}\end{matrix}\ \vec{\psi}_n$$

We observe that the coefficients of expansion A_n^i and F_n^i associated with the exciting and scattered fields depend on the position of all N scatterers. Further, since Eq. (3) is satisfied, we can relate the two sets of coefficients by means of the T-matrix as defined by Waterman[10]. We have

$$F_n^i = \sum_m T^i_{nm} A^i_n \tag{9}$$

The T-matrix depends on the frequency of the wave exciting the scatterer as well as its geometry and material properties.

If Eqs. (4), (5) and (9) are substituted in Eq. (2), we would need the translation addition theorems for the vector spherical functions in order that we may refer all expansions in Eq. (2) to a common origin. In compact form

$$\text{Ou}\ \vec{\psi}_n(\vec{\rho}_j) = \begin{cases} \sum_{n'} \sigma_{nn'}(\vec{r}_{ij})\ \text{Re}\ \vec{\psi}_{n'}(\vec{\rho}_i) ; & |\vec{r}_{ij}| > \vec{\rho}_i \\ \sum_{n'} R_{nn'}(\vec{r}_{ij})\ \text{Ou}\ \vec{\psi}_{n'}(\vec{\rho}_i) ; & \vec{\rho}_i > |\vec{r}_{ij}| \end{cases} \tag{10}$$

where $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$ is the vector connecting O_j to O_i and $\sigma_{nn'}$ is the translation matrix for the vector functions and $R_{nn'}$ is the same as $\sigma_{nn'}$ with the spherical Hankel functions in $\sigma_{nn'}$ replaced by spherical Bessel functions. Detailed expressions for the matrices are given by Boström[11].

The incident electric field $\vec{E}^o$ can be expanded with respect to an origin at O_i as

$$\vec{E}^o(\vec{r}) = e^{ikz} = e^{ik\hat{z}\cdot\vec{r}_i} \sum_n a_n\ \text{Re}\ \vec{\psi}_n(\rho_i) \tag{11}$$

where the coefficients a_n are known (see for example Morse and Feshbach[12]). We observe that for a plane wave propagating in the z-direction the only non-zero values of $a_n = a_{\tau\ell m\sigma}$ are $a_{1\ell 1 1}$ and $a_{2\ell 1 1}$; $\ell \epsilon [1,\infty]$, all other coefficients being zero.

Using Eqs. (4),(5),(9)-(11) in Eq. (2), using the orthogonality of the vector spherical functions we obtain

$$A_n^i = e^{ik\hat{z}\cdot\vec{r}_i} a_n + \sum_{j\neq i}^{N} \sum_{n'} \sum_{n''} \sigma_{n'n}(\vec{r}_{ij})\, T^j_{n'n}\, A^j_{n''} \qquad (12)$$

Equation (12) is a set of coupled algebraic equations for the exciting field coefficients associated with each scatterer. If the number of scatterers N is finite and the position of all the scatterers is known, then Eq. (12) can be solved in principle. But we wish to consider the case $N\rightarrow\infty$, $V\rightarrow\infty$ such that $N/V = n_0$ is a finite number density. Since N is large, we are only interested in the configurational average of Eq. (12) over the positions of all particles

<u>The coherent field</u>

The average of Eq. (12) over the position of all scatterers (the average exciting field) is the same as the ensemble average, where the ensemble is composed of different possible configurations of the scatterers. Equation (12) is averaged over the position of all particles except the i-th. But the right hand side of Eq. (12) explicitly depends on the position of the j-th particle. Hence we must specify the two particle joint probability density $P(\vec{r}_j|\vec{r}_i)$. Further, we assume that all scatterers are identical, so that

$$\left\langle A_n^i \right\rangle_i = e^{ik\hat{z}\cdot\vec{r}_i} a_n + (N-1) \sum_{n'} \sum_{n''} T_{n'n''} \int_V P(\vec{r}_j|\vec{r}_i)\, \sigma_{n'n}(\vec{r}_{ij}) \left\langle A^j_{n''} \right\rangle_{ij} d\vec{r}_j \qquad (13)$$

We note that the average exciting field with one scatterer held fixed is given in terms of the average with two scatterers held fixed, leading to a heirarchy that requires knowledge of higher order probability densities. It has been customary to truncate the heirarchy by invoking the 'quasi crystalline approximation' (QCA) first introduced by Lax[2]. According to this approximation

$$\left\langle A^j_{n''} \right\rangle_{ij} \simeq \left\langle A^j_{n''} \right\rangle_j \qquad (14)$$

Specifically the QCA neglects multiple scattering between pairs of scatterers. Improvements to the QCA have been suggested by Twersky[1] and in previous work by us[5].

The joint probability density is defined as

$$P(\vec{r}_j|\vec{r}_i) = \begin{cases} \frac{1}{V}\, g(|\vec{r}_j-\vec{r}_i|) \;;\; |\vec{r}_j-\vec{r}_i| \geq 2a \\ 0 \qquad\qquad\qquad ;\; |\vec{r}_j-\vec{r}_i| < 2a \end{cases} \qquad (15)$$

Equation (15) implies that the particles are hard (no-interpenetration) and the excluded volume is a sphere of radius 'a' although the particles themselves may be non-spherical. The function $g(\vec{r}_i - \vec{r}_j)$ is called the pair correlation function and depends only on $\vec{r}_j - \vec{r}_i$ due to translational invariance of the system under consideration.

We assume that the coherent field propagates in the same direction as the incident field with a new, effective wavenumber K that is complex and frequency dependent. Hence

$$\left\langle \vec{E}_i^e(\vec{r}) \right\rangle_i = A\, e^{iK\hat{z}\cdot\vec{r}} \qquad (16)$$

where A is the amplitude of the coherent wave. Thus the average exciting field coefficient

$$\left\langle A_n^i \right\rangle_i = \left\langle A_{\tau lm\sigma}^i \right\rangle_i = e^{iK\hat{z}\cdot\vec{r}_i}\, X_{\tau lm\sigma}\, \delta_{m1} \left[\delta_{\tau 1}\delta_{\sigma 2} + \delta_{\tau 2}\delta_{\sigma 1}\right] \qquad (17)$$

The Kronecker deltas in Eq. (17) indicate that only the azimuthal index m=1 contributes, since the coherent wave propagates in the z-direction and those in the square bracket indicate that there is no depolarization.

Equations (14)-(17) are substituted in Eq. (13). Since the T-matrix of a rotational symmetric scatterer is block diagonal in the azimuthal index (see Waterman[10]) and the coherent field propagates in the z-direction, the sums associated with the azimuthal indices of the super indices n' and n" in Eq. (13) are removed. Further, as shown in previous work by us[5] as well as Twersky[7], the extinction theorem can be used to cancel the incident wave term in Eq. (13) with the contribution of the integral at infinity. Finally Eq. (13) can be written in the form

$$X_{\tau lm\sigma}\,\delta_{m1}\left[\delta_{\tau 1}\delta_{\sigma 2} + \delta_{\tau 2}\delta_{\sigma 1}\right] = \frac{N-1}{V} \sum_{\tau' l' \sigma'} \sum_{\tau'' l'' \sigma''} T_{\tau' l' 1\sigma', \tau'' l'' 1\sigma''} \cdot$$

$$\sum_{\lambda=|l-l'|}^{l+l'} I(K,k,c,\lambda)\, X_{\tau'' l'' 1\sigma''}\, D^{(\lambda)}_{\tau' l' 1\sigma', \tau l 1\sigma} \qquad (18)$$

where

$$I(K,k,c,\lambda) = \frac{6c}{(Ka)^2-(ka)^2}\left[(2ka)\, j_\lambda(2Ka)\, h_\lambda'(2ka) - 2Ka\, h_\lambda(2ka)\, j_\lambda'(2Ka)\right] +$$

$$24c \int_1^{\infty} x^2\left[g(x)-1\right] h_\lambda(kx)\, j_\lambda(Kx)\, dx \qquad (19)$$

and

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$$D^{(\lambda)}_{\tau'\ell'1\sigma',\tau\ell1\sigma} = i^{\ell-\ell'+\lambda} \left[\frac{(2\lambda+1)(2\ell+1)}{2\ell(\ell+1)}\right] \left[\frac{\ell'(\ell'+1)}{\ell(\ell+1)}\right]^{1/2} \begin{bmatrix} \ell & \ell' & \lambda \\ 1 & -1 & 0 \end{bmatrix} \cdot$$

$$\left\{ \begin{bmatrix} \ell & \ell' & \lambda \\ 0 & 0 & 0 \end{bmatrix} [\ell'(\ell'+1) + \ell(\ell+1) - \lambda(\lambda+1)]\, \delta_{\tau\tau'}\, \delta_{\sigma\sigma'} + i \begin{bmatrix} \ell' & \ell & \lambda-1 \\ 0 & 0 & 0 \end{bmatrix} \cdot \right.$$

$$\left. (\lambda^2-(\ell'-\ell)^2) \quad (\ell'+\ell+1)^2-\lambda^2) \quad ^{1/2} (1-\delta_{\tau\tau'})\, (\delta_{\sigma'1}\delta_{\sigma 2} - \delta_{\sigma'2}\delta_{\sigma 1}) \right\} \quad (20)$$

In Eq. (19) $c = \frac{4\pi}{3} n_o a^3$ is the effective spherical concentration of the particles and in Eq. (20) $\begin{bmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{bmatrix}$ is the Wigner 3-j symbol.

If the integral in Eq. (19) can be evaluated for suitable models of the pair correlation function, then Eq. (18) is a set of coupled, homogeneous, algebraic equations for the coherent field expansion coefficients. For a non-trivial solution, the determinant of the coefficient matrix must vanish. This yields the required dispersion equation for the effective or average medium. In general the system of equations can be solved only numerically to yield the effective wave number K as a function of frequency ($k=\omega/c$) which is complex ($K = K_1+iK_2$). The real part K_1 yields the phase velocity in the medium and the imaginary part K_2 leads to damping of a propagating wave due to geometric dispersion as well as real losses if any, associated with the discrete particles. We now proceed to consider the evaluation of the integral in Eq. (19).

The Percus-Yevick pair correlation function

The pair correlation function for an ensemble of particles depends on the nature and range of the interparticle forces. The average of several measurements of a statistical variable that characterizes an ensemble will depend on the pair correlation function. As we have seen, the coherent or average electric field in an ensemble of dielectric scatterers depends on the pair correlation function (Eqs. (18)-(19)). To obtain expressions for the pair correlation function, one needs a description of the interparticle forces. In our case we assume that the dielectric scatterers behave like effective hard spheres (where the radius 'a' is that of the sphere circumscribing the scatterer). Percus and Yevick[7] have obtained an approximate integral equation for the pair correlation function of a classical fluid in equilibrium. Wertheim[8] has obtained a series solution of the integral equation for an ensemble of hard spheres. The statistics of the fluid are then same as those of the ensemble of discrete hard particles that we are considering.

Although integral expressions for the correlation functions also result in a heirarchy, Percus and Yevick have truncated the heirarchy by making certain approximations that result in a self-consistent relation between the pair correlation function $g(x)$ and the direct correlation function $C(x)$. The direct correlation

function may be interpreted as the correlation function resulting from an 'external potential' that produces a simultaneous density fluctuation at a point and the external potential is taken to be the potential seen by a particle given that there is a particle fixed at another site. Fisher[13] comments that the Percus-Yevick approximation is a strong statement of the extremely short range nature of the direct correlation function. The integral equation has the form

$$\tau(x) = 1 + n_o \int_{x<2a} \tau(x')dx' - n_o \int_{\substack{x'<2a \\ |x-x'|>2a}} \tau(x')\tau(x-x')dx' \tag{21}$$

where

$$\begin{aligned} \tau(x) &= g(x) \; ; \; x > 2a \\ g(x) &= 0 \; ; \; x < 2a \\ \tau(x) &= -C(x) \; ; \; x < 2a \\ C(x) &= 0 \; ; \; x > 2a \end{aligned} \tag{22}$$

Wertheim[8] has solved the integral equation by Laplace transformation that results in an analytic expression for C(n) in the form

$$C(x) = -(1-\eta)^{-4} \left[(1+2\eta)^2 - 6\eta(1+\tfrac{1}{2}\eta)^2 x + \eta(1+2\eta)^2 x^3/2\right] \; ; \; \eta = c/8 \tag{23}$$

where *'c' is the effective spherical concentration of the particles.* The Percus-Yevick approximation fails as the concentration approaches the close packing factor for spheres and is expected to be good for $c < 0.3$ or 0.4.

Equation (23) can be substituted back into Eq. (21) to yield a series solution for g(x) in the form[8]

$$g(x) = \sum_{n=1}^{\infty} g_n(x) \tag{24}$$

where

$$g_n(x) = \frac{1}{24\eta x i} \int e^{t(x-n)} \left[L(t) \mid S(t)\right]^n t dt \tag{25}$$

where

$$S(t) = (1-\eta^2)t^3 + 6\eta(1-\eta)t^2 + 18\eta^2 t - 12\eta(1+2\eta) \tag{26}$$

and

$$L(t) = 12\eta \left[(1+\eta 2)t + (1+2\eta)\right]. \tag{27}$$

Throop and Bearman[9] have tabulated $g(x)$ as a function of x for values of $\eta = c/8$. A few representative plots of the pair correlation function are shown in Fig. 2. These tabulated values were used in evaluating the integral in Eq. (19).

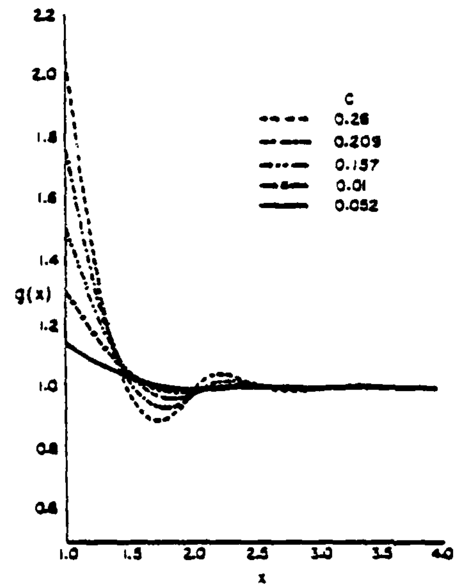

Figure 2. The Percus-Yevick pair correlation function g(x)

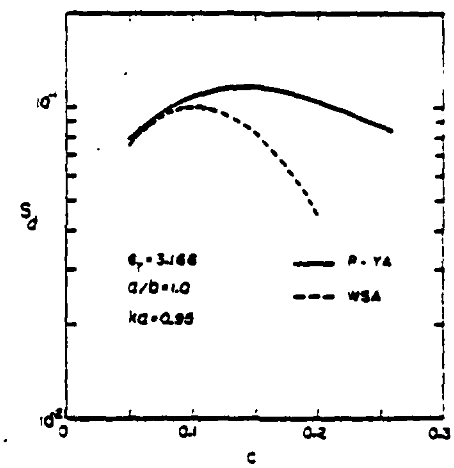

Figure 3. Coherent attenuation vs. concentration for spherical ice particles

Comparison of WSA and P-YA

The homogeneous system of algebraic equations for the effective exciting field were solved numerically for two different models of the correlation integral I appearing in Eq. (18). In eq. (19) if the second term is set equal to zero, we just have a system of uncorrelated hard particles. This is what we have referred to as the well stirred approximation (WSA)[6] earlier. Computations were also performed by numerically evaluating the integral in eq. (19) by using the tabulated values of the Percus-Yevick approximation to the pair correlation functions provided by Throop and Beerman[9].

In Fig. 3, the specific damping $S_d = 4\pi K_2/K_1$ is plotted as a function of concentration for a random distribution of numerical ice particles ($\epsilon_r = 3.168$) in free space at ka = 0.55. The WSA agrees with the P-YA solution only up to concentrations $C \backsim 0.075$ and then there is a marked difference and the WSA fails completely at $C > 0.125$ leading to unphysical results. In Fig. 4, the calculations are repeated

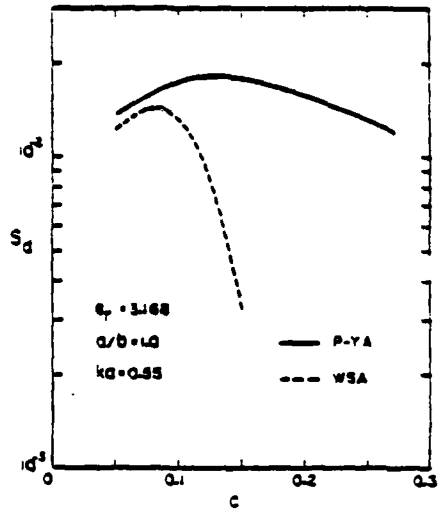

Figure 4. Coherent attenuation vs. concentration for spherical ice particles

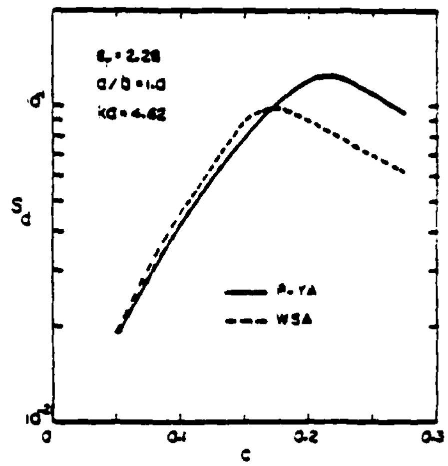

Figure 5. Coherent attenuation vs. concentration for polyethylene spheres

for the same system at a higher value of ka = 0.95. Here the WSA agrees with P-YA up to C = 0.1 and in Fig. 5 similar calculations were performed for polyethylene spheres (ϵ_r = 2.26) at ka = 4.62. For this case WSA and P-YA results agree up to C = 0.15. From these results it would appear that although the WSA is very poor at higher scatterer concentrations, the results improve dramatically at higher values of ka, yielding reasonably good results for higher concentrations. The natural explanation is that at higher values of ka, multiple scattering effects between pairs of particles become smaller and thus pair correlation effects are not significant and the QCA also becomes more exact. But for arbitrary concentration and frequency it is safer to use the Percus-Yevick approximation.

The effective dielectric constant

Once the effective complex wavenumber K has been computed by solving Eq. (18) numerically, we can proceed further and evaluate the effective dielectric constant of the medium which is also complex and frequency dependent. In the usual way, the dielectric constant $\epsilon_r^*(\omega)$ of the random medium is defined as

$$\epsilon_r^*(\omega) = \frac{K^2}{k^2} = \epsilon_1(\omega) + i\epsilon_2(\omega)$$

where ϵ_1 and ϵ_2 are the real and imaginary parts of the dielectric constant and the

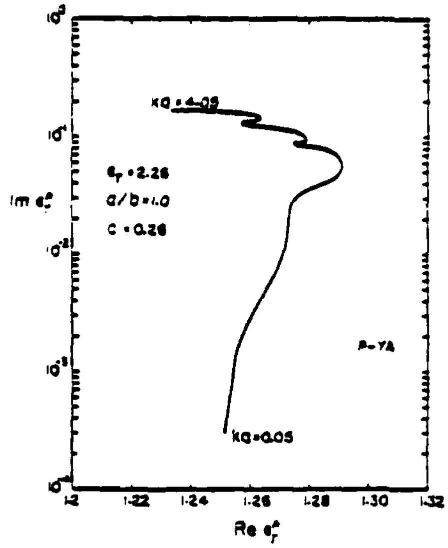

Figure 6. Complex plane locus of the effective dielectric constant for a system of polyethylene spheres

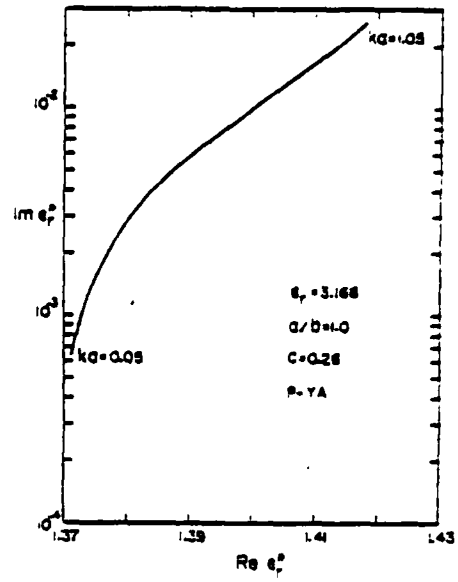

Figure 7. Complex plane locus of the effective dielectric constant for a system of spherical ice particles

subscript on ϵ_r^* denotes 'relative to the matrix medium'. The real part ϵ_1 is related to the refractive index and phase velocity in the artifical medium and the imaginary part ϵ_2 accounts for the damping in the medium. In real materials, the damping is intrinsic to the system and is due to macroscopic viscosity of the dielectric. For the artificial or effective medium under consideration, in addition to natural losses there is damping due to geometric dispersion or scattering.

Cole and Cole[14] have given a convenient representation of the dispersion and absorption in a dielectric by means of an Argand diagram or a plot in the complex ϵ-plane of ϵ_1 versus ϵ_2, each point of the plot being characteristic of a particular frequency. For many types of loss mechanisms, the locus of the points is a semi circle with its center on the real axis or a circular arc. In Ref. 14, the complex dielectric constant of several liquids and solids is plotted conforming to the circular arc.

In the present case the complex dielectric constant $\epsilon(\omega)$ corresponding to the effective wavenumber K of the effective medium is studied for several values of the frequency. Overall results show a dramatic deviation from the circular arc locus. This is to be expected since the medium is artificial.

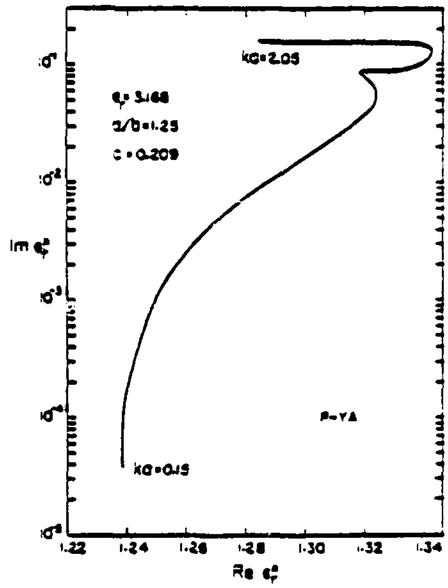

Figure 8. Complex plane locus of the effective dielectric constant for a system of oblate spheroidal ice particles

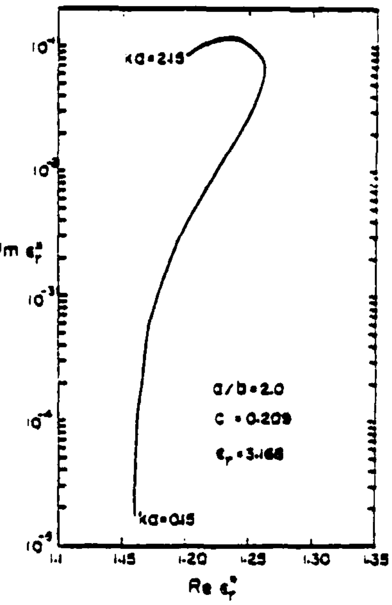

Figure 9. Complex plane locus of the effective dielectric constant for a system of oblate spheroidal ice particles

In Fig. 6 the complex plane locus of the relative dielectric constant of a random distribution of polyethylene spheres in free space is presented at a concentration of 26%. The calculations were done using the Percus-Yevick approximation (P-YA) for the pair correlation function from ka = 0.05 to 4.05. As can be seen, the figure bears no resemblance to a circular arc locus. By extrapolating the locus at the low value of ka, one can find the intercept on the Re ϵ_r^* axis which is equal to the static dielectric constant of the effective medium. Since the dielectric constant of the spherical particles is assumed to be real, the effective medium shows no absorption at low frequencies. The static dielectric constant thus obtained will correspond to the one that can be obtained from mixture theory. In real media displaying a circular arc locus the high frequency value of ϵ^* also intercepts the real axis and this yields the optical limit or ϵ_∞ for the material. In our case, it is not at all clear at what value of ka, if at all, the locus will intercept the real axes.

In Figs. 7,8 and 9 the complex plane locus of the effective dielectric constant of spherical and oblate spheroidal ice particles is presented where 'a' and 'b' are the semi major and semi minor axes respectively. They all show marked deviation from the circular arc locus and it is unclear what ϵ_∞ will be for these effective media.

At the present time there are no experimental results available to verify these calculations. The practical applications of these computations are many. Such calculations will provide reasonable estimates of the frequency dependence dielectric constant as a function of particle concentration, size and shape for inhomogeneous media.

Acknowledgements

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